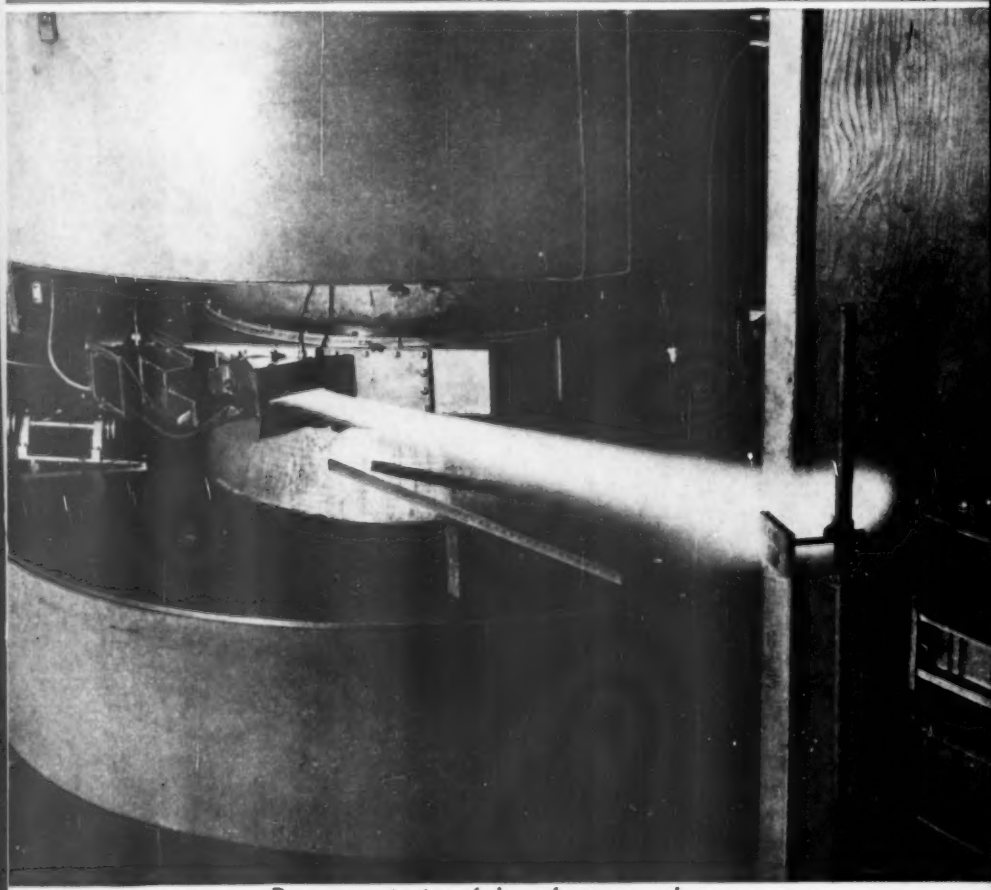
NUCLEONICS

TECHNIQUES AND APPLICATIONS OF NUCLEAR SCIENCE AND ENGINEERING

October, 1951



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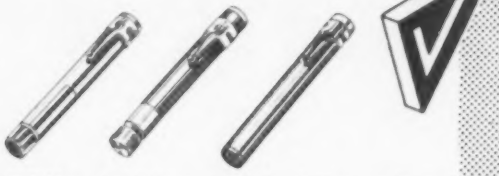

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NUCLEONICS

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THE COVER shows how samples of glass refractories were activated in the deuteron beam of the cyclotron at the University of Pittsburgh. Autoradiography was then used to study the effects molten glass had had upon the specimens. The samples were placed near the end of the beam where the particle energy was about 4 Mev, as described on page 44

Vol. 9, No. 4

Radioisotopes Techniques Conference—Oxford, 1951... 5
J. L. PUTMAN

Calculation of Yields in Multiple Neutron Capture Processes... 17
R. F. VAN WYE and J. G. BECKERLEY

Relative Sensitivities of Windowless and End-Window Counters... 22
WALTER L. GRAF, C. L. COMAR and IRA B. WHITNEY

A New High-Resolution System of Autoradiography... 28
HENRY J. GOMBERG

Deuteron Activation and Autoradiography as Coupled Techniques in Tracer Experiments... 44
K. M. LAING, R. E. JONES, D. E. EMHISER, J. V. FITZGERALD and G. S. BACHMAN

Detection of Small Fluid Flows by Means of P^{32}... 47
F. T. ROGERS, Jr. and L. E. SCHILBERG

The Linear Electron Accelerator as a Pulsed Neutron Source... 51
B. T. FELD

Nomogram for Estimating Decay of I^{131} and P^{32}... 58
WILLIAM B. MILLER, Jr.

Instrumentation and Control of Reactors... 60
J. D. TRIMMER and W. H. JORDAN

Departments

EDITORIAL 2
BOOKS 69
CROSS SECTIONS 72
NUCLEONIC EVENTS 78
PRODUCTS AND MATERIALS 84

A Reexamination of the Plastic Crystal Ball

In New York last month, Dr. James B. Conant, president of Harvard University, looked into his crystal ball—a plastic one in honor of the diamond jubilee meetings of the American Chemical Society—and predicted a peaceful conclusion to the 20th century. And with peace, he foresees the dawn of a new energy era, one in which solar energy far overshadows nuclear energy. We hope he is right in his first prediction but question the details of the second.

Peace is possible because atomic warfare will never take place, according to Dr. Conant. He feels that the potential military applications of atomic energy are "inherently inimical to the very nations that control the weapons." Atomic weapons will never be used, he says, because military experts feel that they cannot "guarantee ultimate success."

He suggests that, because of a military stalemate, international control of atomic weapons will become possible in 15–20 years and that existing stocks of fissionable materials will be put beyond the immediate reach of any nation.

In part because of these military factors, Dr. Conant states further that, by the 1960's, "a sober appraisal of the debits and credits of the exploitation of atomic fission . . . led people to decide the game was not worth the candle."

But also included in his reasoning behind this pessimistic prediction is his feeling that the problems of the disposal of radioactive wastes and the great capital investments necessary for the development of nuclear power are seemingly insuperable.

On examining Dr. Conant's comments closely, we are left with mixed feelings. On the one hand, we sincerely hope that his optimism about international peace is borne out. But on the other, although we do wish that he is

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correct in his statement that a valuable new energy source is on the horizon, we question his prediction.

Our crystal ball does not give us the same picture which Dr. Conant's gave him. He feels that by the end of this century solar energy is going to be the dominant energy source. Our crystal ball gets quite cloudy here—discolored, perhaps, by nuclear and/or solar radiation. We can't get a clear enough picture to predict the future of either nuclear or solar energy.

This is our main point of disagreement with Dr. Conant. We don't think that he—or anyone else—has enough information at this time to make a valid prediction on this question.

However, we must consider Dr. Conant's statements very seriously. With an outstanding record in the fields of science and education, he is not prone to making rash predictions. More important, as a member of the General Advisory Committee of the U. S. Atomic Energy Commission, he is as close to scientific developments in the field of atomic energy as anyone might be.

In general, we do not question the right of anyone to make predictions. Frequently they make for light and enjoyable reading. However, because of Dr. Conant's closeness to this field, especially his position on the GAC, we expected that he would have had more information on this subject than is available to the general public. Actually this is not the case, as we learned in conversation with him following his talk.

Let's look at the facts of the situation.

Energy Needs of the World

There seems to be fairly common agreement that within the next 25–50 years there is going to be the need for the development of a significant new source of low-cost energy. Our present energy reserves will not be able to keep up with the energy needs of growing population and a growing industrial economy.

It has been estimated* that in the next 50 years the world population may jump from about 2.3 billion to 3.8 billion, an increase of about 70%.

In the 1,850 years to the year 1850, the world used energy totaling $6–9 \times 10^{18}$ BTU. In the following century (to 1950), about 4×10^{18} BTU were used. In the next 50 years we may require $10–20 \times 10^{18}$ BTU.

World energy reserves (exclusive of nuclear and solar energy), economically recoverable at substantially present costs, add up to about 30×10^{18} BTU.

The need for new energy sources is thus quite apparent.

How do nuclear and solar energy fit into the picture?

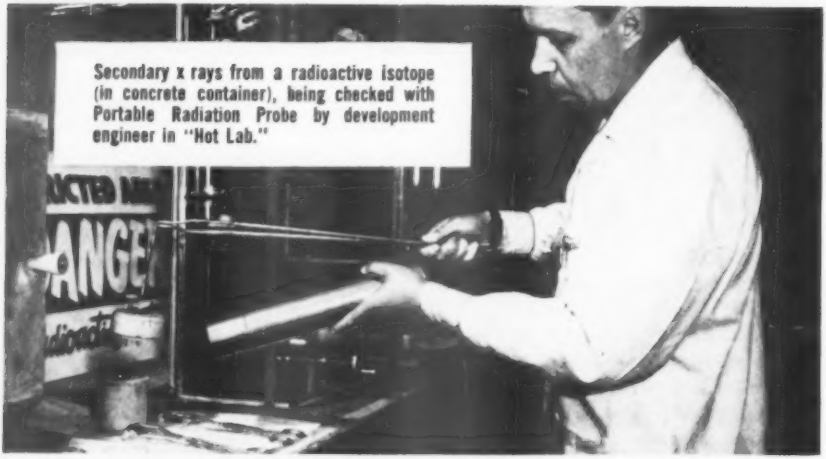
Solar Energy

Much less is known today about the technical aspects of harnessing solar energy than is known about atomic energy. Early this year, a U. S. Senate

* Data given here on population growth, on world energy needs and later on energy available from fissionable material were presented by Palmer C. Putnam, consultant to the AEC, in a series of open lectures at Oak Ridge.

(*Continued on page 88*)

How the NEW G-E PROBE detects thermal Neutrons and Alpha, Beta, Gamma Radiation



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Radioisotopes Techniques Conference — Oxford, 1951

This is a report, prepared especially for NUCLEONICS, on the British-sponsored meetings to discuss radioisotopes techniques and applications in medicine, agriculture, and industry. Arranged by the Atomic Energy Research Establishment at Harwell, England, the conference took place at Oxford from July 16–20, 1951 and was attended by 450 delegates from all over the world. Thirty sessions comprised the program, and ninety-eight papers were presented and discussed. The conference was accompanied by an exhibition of isotope techniques in the Clarendon Laboratory, Oxford

By J. L. PUTMAN
Isotope Division, Atomic Energy Research Establishment
Harwell, England

THE FIRST TWO DAYS of the Radioisotopes Techniques Conference were devoted to medical and biochemical applications. The third day was divided between biochemical and agricultural applications, and the last two days were devoted to industrial applications and related research.

An attempt has been made to report selected sessions, together with an impression of about fifty of the ninety-eight papers presented.*

Medical and Biochemical Applications

Many excellent papers were presented in the medical and biochemical fields. They have been grouped below under appropriate subheadings for ease in reference.

Therapy and Diagnosis

The first lecture was given by *Prof. W. V. Mayneord* (London, England) who discussed problems of deducing the distribution of radioactive material in the body from the distribution of dosage rate or counting rate near the source.

Equipment was described which gives visual indication on a cathode-ray oscilloscope of the distribution of counting rate over an area, thus eliminating laborious plotting of the distribution from point observations. Two symmetrical screened gamma counters are used, one being collimated to accept a narrow beam of radiation and the other being used for background corrections. The device is made to scan the required area as indicated by a beam of light coming through the collimator. Counting rates, integrated over half-minute periods, are made to modulate the brightness of an oscilloscope spot which scans in synchronism with the collimator. A visual picture is obtained that can be observed directly, using a cathode-ray tube with a long afterglow, or recorded photographically, using a vivid screen.

Preliminary tests with known artificial distributions indicate that resolutions are in accordance with optical theory.

The uses of P^{32} in internal therapy

* Proceedings of the conference, including papers in full, and the subsequent discussions, will be published shortly by H. M. Stationery Office, London, and will be on sale to the public. They will comprise two volumes: Vol. 1 will include the medical, biochemical and agricultural papers; Vol. 2 will deal mainly with industrial and allied research applications.

Next month NUCLEONICS will start publishing selected papers from the Radioisotopes Techniques Conference—Oxford, 1951. Among these will be:

Some Industrial Applications of Radioelements Made at the Commisariat a L'Energie Atomique (Jules Gueron)

The Measurement and Handling of Radioactive Materials (W. K. Sinclair)

The Design of Directional Counters for Clinical Use (Brian D. Corbett and A. J. Honour)

A Study of Tracer Methods for Assessing the Wear of Wire-drawing Dies (J. C. E. Button, A. H. Davies, and R. Tourret)

Estimation of Tritium and Some Preliminary Experiments on Its Use as a Label for Water (R. F. Glascock)

and for external applicators were described, and considerable discussion took place on the therapeutic and diagnostic uses of I^{131}. Teletherapy units using Co^{60} and Ir^{192} were also described.

Dr. J. W. Otte (Madrid, Spain) described the use of methionine, cysteine, and hydroquinones to increase the uptake of P^{32} and to increase the general tolerance in the treatment of melanomata with multiple metastases and of bone metastases of breast carcinoma.

Dr. A. Lindblom (Växjö, Sweden), describing the treatment of leukemia with P^{32}, distinguished between chronic lymphatic leukemias and chronic myelotic leukemias as showing different response to P^{32} therapy. In the lymphatic leukemias, notable remissions and sometimes complete disappearance of symptoms were obtained, but in the myelotic leukemias dosage was limited by poor uptake in the spleen.

In chronic lymphatic leukemias, P^{32} gives much better results than local X-ray therapy. Large spaced doses of 0.08 to 0.12 mc per kilo of body weight have been found more successful than small frequent doses, and careful control of the thrombocytes is important. For oral administration, 20–25% higher dose is required than for intravenous injection.

Dr. B. Chr. Christensen and *Dr. H. Bohr* (Copenhagen, Denmark) reported on the treatment of polycythemia vera with P^{32} and found that patients fell into two groups, one showing both clinical hematological improvement and reduction of the circulatory blood volume after treatment, and one showing neither. It was concluded that two varieties of the disease exist, one being resistant to P^{32} treatment.

Dr. L. Leksell and *Dr. K. Liden* (Lund, Sweden) described a stereotaxic method for the application of radioisotopes in brain tumors and reported its use with P^{32} in a single case of cystic craniopharyngeoma in which some improvement has resulted.

Dr. A. Bru (Toulouse, France) gave an account of the treatment of Sticker's sarcoma in dogs. In this disease, surgical excision of tumors is frequently followed by relapse, but radiotherapy in doses from 400 to 600 r per tumor is effective. X-radiation or internal doses of P^{32} may be used. P^{32} is deposited selectively in the tumors, where it produces a general regression of the tumor. Histological changes in the tumorous cells produced by P^{32} differ from those produced by X-rays, and destruction is slower with P^{32}.

The design and use of beta-particle applicators was described by *Dr. H. F. Freundlich* (Cambridge, England) and by *Dr. J. Th. van der Werff* (Nijmegen, Netherlands)

Dr. Freundlich described a simple applicator in which P^{32} is deposited on blotting paper and enclosed in cellophane (Low-Beer method). This

method is used in particular for the treatment of nevi in children and superficial lesions, in which its ease of application is a great advantage. Central dose curves were shown for P^{32} and Pr^{144} beta particles, measured with small condenser chambers in a phantom. The use of hard beta particles from Ce^{144}—Pr^{144} and of Ru^{106}—Rh^{106} to produce increased depth doses was proposed.

Dr. van der Werff described a method suitable for the local application of P^{32} to small inaccessible tumors. P^{32} in the insoluble form of zirconium phosphate is prepared in suspension in peanut oil, with specific activity 1 mc/ml. The suspension flows freely when warm and is easily injected; after injection, however, it remains stationary in tissue. Two successful applications were described, one to a subcutaneous carcinoma of the lower eyelid and one, made during operation, to a tumor of the bladder. A plastic applicator composed of zirconium phosphate in paraffin wax is in an early stage of trial and has had limited success.

I^{131} Therapy for Thyroid Cancer

Dr. Ralston Paterson (Manchester, England) discussed the selection of tumor types that selectively absorb iodine. These comprised approximately one fifth of the cases studied. Preliminary tracer studies of each patient are made with I^{131} before therapy. Direct measurement of activity is possible in metastases remote from the thyroid, and autoradiography can be used with small excised portions of the tumor. For treatment, high doses up to the limit of whole-body radiation at three-month intervals are favored over frequent low doses. When small doses are used, destruction of thyroid tissue by earlier doses is likely to reduce subsequent uptake by the tumor.

In the experience of *Dr. J. Nielsen* (Copenhagen), although operation is at present the treatment of choice for carcinoma of the thyroid, radioiodine therapy should be tried in all inoperable cases. This treatment should be refused only when the iodine uptake of metastases has definitely been proved zero, and the only adequate test is the administration of a therapeutic dose. Evidence of slight uptake should be followed, in Dr. Nielsen's opinion, by thyroidectomy to increase the uptake in metastases, and large therapeutic doses of I^{131} should then be given. Dr. Nielsen observed the danger that, in some thyrotoxic carcinomas, treatment with I^{131} may cause slowly growing neoplastic tissue to flare, turning a benign tumor into a malignant one.

In all thyrotoxic patients treated with I^{131}, thyrotoxicosis was reduced.

Dr. E. E. Pochin (London) also discussed the selection of cases of thyroid carcinoma for radioiodine treatment. In assessing the likelihood of selective uptake of iodine by cancer tissue, it is probable that a slowly growing tumor, histologically similar to the thyroid, will concentrate iodine. Iodine concentration can be measured if the metastases are well separated from the thyroid.

Autoradiography of excised tissues was discussed as positive evidence of iodine concentration. It was pointed out that negative autoradiographs do not necessarily indicate that metastases are incapable of absorbing iodine, unless it is certain that the test dose is high enough. Test doses as high as 3 mc may be needed.

The use of plane-collimated Geiger counters for measuring body profiles as a diagnostic method was discussed, and the importance of familiarity with the normal pattern of iodine distribution was stressed. Uncertainties exist near the thyroid and liver, which concentrate bound organic iodine. Evidence is often much clearer after thyroidectomy. This is due partly to masking of results

by the strong radiations from the thyroid, partly to competitive uptake of iodine by the thyroid, leaving only small quantities for the tumor, and possibly to stimulation of the tumor to absorb iodine after the removal of the thyroid. In one case quoted, a single dose destroyed one metastasis, and a second dose was concentrated in a different area where metastases had not previously been apparent.

Thyrotoxicosis Diagnosis with I^{131}

Dr. A. Macgregor (Sheffield, England) described various methods in which the metabolic turnover of radioiodine is used clinically as an index of thyroid function.

He described and discussed the relative merits of thyroid uptake and clearance rates and urinary excretion rates of iodine as indices of thyrotoxicosis. In each of these tests, some overlap was found between measurements on clinically thyrotoxic and nontoxic patients, although the use of two or more methods was generally sufficient to establish whether a particular case was thyrotoxic or not.

These workers found that the estimation of protein bound iodine in the plasma is a reliable index of thyrotoxicosis. Routine estimations are made on outpatients 48 hours after the oral administration of a tracer dose. Thyrotoxicosis is indicated if the protein-bound fraction of the administered dose is greater than 0.4% per liter of plasma (186 cases examined). It was found that the protein-bound iodine index may not return to normal after therapy, although other indices may be normal and clinical symptoms may disappear.

Dr. Schiller, presenting a paper with *Dr. Gilbert Dreyfus* (Paris, France) gave an interpretation of the now familiar iodine-uptake curves associated with hyperthyroid, normal, and hypothyroid conditions. He showed, in particular, how in hyperthyroidism both the iodine uptake and thyroxin secretion by the thyroid are abnormally rapid.

The rate of destruction of protein-bound iodine by the tissues is also abnormally rapid, and a single determination of protein-bound iodine at the end of the 24th or 48th hour is inconclusive as a diagnostic test. Measurement of the protein-bound iodine in the plasma in the first hours after dosage is of diagnostic significance, being proportional to the thyroid uptake, provided that the absorbed iodine does not diffuse into the extracellular fluid during that time.

In treatment, the active volume of the thyroid is reduced, influencing both uptake and secretion rates. Hyperthyroid conditions can be reduced more closely to normal by I^{131} than by subtotal thyroidectomy.

Prof. J. Rotblat and *Mr. R. Marcus* (London) gave a mathematical analysis of the distribution of iodine in the body in the first few hours after intravenous injection and showed how the metabolic processes involved would be expected to affect the urinary excretion rate of iodine. Practical curves of urine excretion follow expected shapes in being composed of two exponential functions. Graphical analysis of these curves yields the rate of uptake of iodine by the thyroid. A marked separation was reported between results for normal and disordered thyroid cases (50 cases examined).

Mr. D. G. Arnott (London) described a simple diagnostic arrangement suitable for the study of iodine excretion rates. An iodine-starved patient is given an oral dose containing 5 μc of I^{131}. Urine samples are taken, representing excretion in 0–8 hours, 8–24 hours, and 24–48 hours after administration. Samples are taken in winchesters, then made up to 2 liters with water and assayed for I^{131} by placing them in a ring of 6 vertical gamma counters. The method has been used

1,200 times for diagnosis in the 2 years it has been in use.

I^{131} Therapy for Thyrotoxicosis

Mr. G. W. Blomfield (Sheffield) summarized 30 cases of thyrotoxicosis treated with I^{131}. The treatment was completely successful in 21 cases, with improvement in several others still under treatment. No significant complications were observed. Doses from 5–10 mc administered orally are usually found sufficient. In general, I^{131} therapy is the treatment of choice for thyrotoxicosis in older patients and in all cases where the disease has recurred after operation.

Prof. B. E. Marques (Lisbon, Portugal) outlined the results obtained in I^{131} therapy in several cases of hyperthyroidism. Thyroid uptake, plasma and urine content were checked. Doses from 1–6 mc gave satisfactory results.

Dr. N. Myant (London) discussed the radiation received by various organs during treatment with I^{131}. Timed measurements were made during the various phases following the administration of a tracer dose, and from these the integrated irradiation of the vital organs resulting from a therapeutic dose was calculated. Most cases of damage occurred in organs situated close to the thyroid or bladder, indicating that direct irradiation from iodine concentrated in these places was responsible. Results of animal experiments (e.g., with mice) can be misleading if applied to human cases because of differences in radiation absorption in the different distances between the organs. It was concluded that the hazard to a patient from irradiation of the vital organs in I^{131} therapy is small.

Gamma-ray Therapy

Dr. K. Scheer (Heidelberg, Germany) described the use of a plastic, uniformly impregnated with Co^{60} in powder form. The plastic is cut to required shapes for direct application to a tumor. It is then enclosed in cellophane thick enough to absorb the beta particles. The method is used mainly for superficial and intracavital lesions. The dosage is measured with a dosimeter, but the activity of the applicator can be calculated quickly by weighing after cutting to shape.

Dr. F. Ellis (Oxford, England) showed a model of a proposed teletherapy source which will use Co^{60}. Its total strength will be 2,400 curies, distributed in 30 Co^{60} sources around a circle 200 cm in diameter and arranged to direct the radiation toward the center. Collimating channels operated with mercury will allow control of irradiated volumes from a diameter of 30 cm down to 2 cm. The source is expected to be equivalent to a 2-Mev X-ray apparatus, but with advantages in geometry, flexibility and economy.

Dr. H. F. Freundlich reported on the Ir^{192} teletherapy unit as used with *Prof. J. S. Mitchell* and others at Cambridge. Treatment has been given to 44 cases of mainly cancer of the pharynx, larynx and mouth. The gamma-ray dose required to produce comparable effects is approximately 1.2 times that of a 220-kev X-ray machine. Dose rate is 14–16 r/min. A stronger source is planned.

Dr. Freundlich, quoting the results of Prof. F. W. Spiers, stressed the advantages of using gamma energies above 300 kev where the differential absorption in bone and tissue is less dependent on energy (absorption is ~50% higher in bone than in tissue).

Biochemical Studies

Among papers delivered in sessions on biochemical investigations was one on the metabolic fate of C^{14}-labeled glycine and acetate in humans, presented by *Dr. L. Hellman* (New York). Doses of 200-μc activity were found safe for administration. From 70–80%

of the C^{14} was rapidly excreted, mostly as respiratory CO_2. The application of C^{14}-tracer techniques to studies of the metabolism of hemoglobin, proteins, steroids, uric acid, and CO_2 was described.

Dr. A. S. MacFarlane (London) showed how C^{14} and S^{35} tracers have been used to study the building up and degradation of proteins in blood plasma.

Dr. G. Popjak (London) gave an account of a study of the origin of fetal lipids for which C^{14}, P^{32} and deuterium tracers were used. It was shown that the lipids are synthesized from small molecules within the fetal tissues and not derived from the placenta or the maternal blood.

Prof. A. Wormall (London) described the use of proteins labeled with P^{32}, S^{35} and I^{131} in the study of immunochemistry. As a result of the use of tracer techniques, very small amounts of serological precipitate were necessary for analysis with respect to antigens and antibodies. The action of specific inhibitors was studied quantitatively.

Chromatography

Mr. F. P. W. Winteringham (Slough, England) in a paper on the use of radioisotopes in paper chromatography, pointed out that much smaller activities could be used in this method than in column chromatography. He described a method of measurement of paper strip chromatograms in which the paper is passed in 5-mm steps past a 5-mm aperture under an end-window Geiger counter, obviating errors in cutting equal strips and maintaining a constant geometry. The paper movement is controlled by an escapement, operated by a timing unit which also resets the recorder.

Typical separation curves were shown. In quantitative estimations, changes of sensitivity of the counter over the 5-mm aperture introduce a small error. Self-absorption causes an apparent drop in activity to 36.6% when using C^{14}, which leads to an indeterminacy of 7.4%. With P^{32}, the reduction is only to 95.7%, and the accuracy of correction is ½%.

Three radioactivity methods were discussed: (1) The mixture to be examined is labeled with radioisotopes before chromatography. (2) Labeled reagents are used after chromatography. (3) A finished chromatogram is analyzed by radioactivation. Here sensitivity is limited by the background activity of the paper.

Dr. R. Michel and *Professor J. Roche* (Paris) described applications of paper chromatography and silica column chromatography in studies of the fate of iodinated amino acids in the thyroid gland. Mono- and diiodotyrosine were identified, and traces of monoiodohistidine and diiodothyroxin were established. By selective labeling of mono- and diiodotyrosine, these authors have shown that the iodotyrosines are deiodinated by an enzyme not acting on thyroxin and that the iodide formed is used again by the thyroid gland.

Dr. J. Sternberg (Montreal), with *Dr. H. Simonnet* (Paris), have used an aluminum oxide column for chromatographic separation of P^{32}-labeled constituents of milk. Rat's milk (3–5 cm^3) is used for a separation. A thin-wall counter at the base of the column continuously records the specific activity of the eluted fraction. The yield is quantitative, and the sensitivity about 100 times that of classical methods. An exchange reaction occurs between mineral and protein-bound phosphorus. This is at present less than 2% but can be reduced by more rapid elution of the protein phosphorus. Experiments are being extended to other phosphorus-containing constituents of milk.

Electron-track Radiography

Dr. D. T. King (Bristol, England) dealt with the technique of electron-

track radiography for tracer investigations in microbiology. In work with *Prof. J. E. Harris*, he has investigated the uptake of phosphors in protozoa, using P^{32} in a nutrient medium. Much lower levels of activity are detectable with the electron-track-radiography method than with a Geiger counter; factors of 20 to 100 in the threshold of detection were quoted. Organisms are mounted alternatively above, below, or within the photographic emulsion, and exposures are of the order of 24 hours. Location of sources is within 5 microns.

Measurements with small cubic crystals of $CaP^{32}O_4$, which could be checked in known numbers with a Geiger counter, established that track counting is an accurate method of assay. Biological losses of P^{32} on fixation (with formalin), dehydration, and treatment with benzole, unfortunately vitiate absolute measurements in the experiments described. It has been established, however, that the absorption of P^{32} is closely associated with the ingestion of food, since starved cultures take up more P^{32} than those that have previously been fed. Phosphorus is found in food vacules, a few microns in cross section. A wide range of uptakes is found, but these appear to be unrelated to either the length of the organism or the phase in the 24-hour life cycle.

Agricultural Applications

The study of soils and fertilizers and of plant nutritional experiments was pursued at some length.

Soils and Fertilizers

Dr. J. Govaerts (Liège, Belgium) described a number of experiments in which the distribution of fertilizers was studied by injecting P^{32}-labeled phosphate solutions below the surface of the ground and assaying samples of soil at intervals after the injection.

Plant Nutrition

Dr. R. Scott Russell (Oxford) surveyed the problem of radiation injury in plant nutritional experiments with P^{32}. He illustrated with autoradiographs that absorbed phosphorus is concentrated in the root tips and stem apex of growing plants.

In soil culture, the factors that decide the permissible level of activity are complicated. Different rates of root growth occur at different seasons, and deeper roots absorb mainly inactive soil phosphorus. Damage is significantly greater in wet than in dry weather. Even in water culture, the effects of radiation are not simple. Damage may be caused by free radical-in the nutrient solution, as well as by direct irradiation.

Radiation damage has even been found on placing a plant in an inactive solution which had previously been irradiated. In studies with barley, radiation damage was found in water-culture experiments involving P^{32} activities above 5 μc/liter and in soil-culture experiments using specific activities of 92 μc/gm of phosphate. In borderline cases, damage has been found after delays up to 56 days.

It is misleading to base assessment of radiation damage on a single criterion, such as shoot weight. Experiments are only valid if no significant injury occurs to affect the processes which are being studied.

Methods of determining the amount of phosphate in the soil which is available to a growing crop were described by *Mr. O. Gunnarsson* (Upsala, Sweden). Pot experiments with wheat indicate that the phosphorus in added superphosphate becomes equally available to plants with plant-available soil

phosphorus. The amount of plant-available phosphorus in the soil can be deduced by measuring the quantity of labeled fertilizer phosphorus as a percentage of the total phosphorus in a harvested plant. Field experiments on similar lines show good general agreement with pot experiments, although it is found that the values vary widely between different soils.

The fact that a wide range of fertilizer quantities added to the soil give the same results in these tests indicates that a dynamic equilibrium between fertilizer phosphate and plant-available soil phosphorus is quickly established. Hence, it is to be expected that the proportions of labeled and unlabeled phosphates in water-soluble form are soon equal to the proportions in the soil.

A laboratory experiment in which phosphates extracted with distilled water were measured, gave results in good agreement with pot and field experiments. The amount of lactate-soluble phosphorus in the soil, as estimated by the chemical extraction method of Egner, is significantly lower.

It was concluded that part of the plant-available phosphorus is dissolved in the soil solution and another part bound to the colloid complex of the soil but in dynamic equilibrium with the soil solution.

Mr. J. B. Rickson (Edinburgh, Scotland) discussed the ion exchange between labeled phosphates added to soil and phosphates already present in the soil. He described laboratory experiments in which P^{32} as orthophosphate was added to a suspension of solid phosphate in a solution with which it was in equilibrium.

Measurements of the rate at which P^{32} is absorbed by the solid show that the absorption is due to two reactions. The first and more rapid process is due to surface adsorption on crystals of the suspended phosphate, followed by some penetration into the crystal surfaces. This effect is proportional to the surface area of the crystal. A slower process involves absorption by recrystallization, due to instability in the crystal lattice. The net absorption may be expressed as the sum of two exponentially decreasing functions of time. Investigations of several natural and synthetic basic calcium phosphates indicate that the crystal size of the synthetic phosphates is much smaller than those of the mineral phosphates.

In the study of plant nutrition, the mechanism of absorption of simple salts has been explored by *Dr. R. E. Davies* and *Miss M. J. Wilkins* (Sheffield). They described the use of potassium bromide double-labeled with K^{42} and Br^{82} to examine the relations between anion and cation absorptions during the uptake of KBr by washed carrot disks. The K^{42} and Br^{82} in the carrot disks and the solutions were measured by counting diluted samples in a liquid counter. The different half-lives of the two isotopes were used to assay the activity of each isotope by using two counts taken at intervals of two or three days.

In general, the potassium uptake was found to be higher than the bromide uptake. Results depended on the salt respiration of the disks and the maximum concentration differences maintained between the disks and the solutions.

Industrial and Research Applications

Under this subject, miscellaneous as well as specific applications of radioisotopes in industry were discussed. The types of application of isotopes in industrial use were outlined by *Dr. H. Seligman* (Harwell) under two main headings.

The first type of application referred

to the use of properties of the radiations, such as ionization in gases and absorption by different thicknesses of material.

The principle of ionization in gases has been used in the design of radioactive static eliminators for electric charges. Low-activity static eliminators consisting of silver foils enclosing Tl^{204} are now commercially available. The design of higher-activity eliminators is being studied at the Atomic Energy Research Establishment at Harwell, and these should be available within a year.

Thickness gages employing the absorption of beta particles and those relying on the back scattering of beta particles were briefly described. The back-scattering gages are capable of measuring tin-plate thickness to 10^{-5} inch, which will soon be reduced to 10^{-6} inch. Reference was made to gamma radiography as essentially a thickness measurement. This subject was discussed very fully in a later session.

The second type of application considered was the use of radioisotopic tracers. Dr. Seligman described examples of this technique, including the use of A^{40} to measure air exchange in a room. He also mentioned the use of Co^{60} to locate a large scraper inside a pipeline and drew attention to the use of tracers to monitor the separation of metals in an ion-exchange column.

Miscellaneous Applications

Dr. J. Guéron (Paris) described some applications of radioisotopes made at the Commissariat à l'Energie Atomique.

He dealt first with problems of leak detection and described the use of Na^{24} in sodium carbonate for pressure tests at 400 atmospheres on pressurized casings for undersea repeaters in telephone systems. The measurement of gas leaks in pressurized underground telephone cables was also described. The cables are normally buried to a depth of 80 cm. The criteria for choosing a suitable tracer gas are that it should not stick to the cable, that it should diffuse easily through the soil, but should remain long enough in the soil to permit measurement. Easily detectable radiations and a half-life of the order of a few days are also desirable. Methyl bromide labeled with Br^{82} was found very suitable for these tests. A build-up time of about one hour after injection of the vapor allows it to diffuse to the surface of the soil, and the spot can be detected for about two days.

Radioactive xenon has been used to test natural and forced ventilations. A radioactive falling ball viscometer was described for measurement of viscosity of opaque viscous fluids. The passage of the ball was detected by Geiger counters behind collimated slips.

On the subject of water tracing in solids by substances dissolved in the water, Dr. Guéron emphasized the errors that may result from adsorption and ion exchange. Experiments are, at present, in a preliminary stage. The elution curve should rise sharply, but, if adsorption occurs, the rise in activity is delayed and a slowly rising threshold followed by a long trailing tail is obtained. Fluorescein is a good label from this point of view. Labeled NaCl and CH_2Br both showed bad adsorption. Bromoform was found very promising.

Dr. R. Cahn (Harwell) gave a review of some uses of radioactive tracers in metallurgy. Among other applications, he described measurements of diffusion in the solid state, especially self-diffusion which is not measurable directly by other means. A Geiger counter with a slit aperture has been used to explore surfaces for measurement of surface diffusion.

Corrosion studies have been approached in two ways. In an example of the first method, a thin plate of radioactive iron on steel is exposed to

corrosion by an inactive solution. Autoradiography of the surface after exposure to the solution shows incipient corrosion, the radiations from the iron being reduced in the oxide. In this method, the quantity of iron in the solution can also be measured. In the alternative method, the solution can be made radioactive and autoradiography is then used to detect radioactive spots on a previously inactive surface.

The distribution of constituents in an alloy can also be examined by autoradiography by using radioactive constituents in the alloy and subsequently taking an autoradiograph. The resolution of this method is limited by the distribution of the radioactive source in depth away from the photographic film. The difficulty can be overcome to some extent by surface activation of the alloy. An alloy is placed in a solution containing one of the constituents as a radioisotope. Ion exchange occurs, resulting in surface activation of the required constituent. This is then followed by autoradiography and the resolution is improved.

Dr. R. Lindner (Göteborg, Sweden) described studies on the reactions of radiotracers on solids. He was able to differentiate between volume diffusion and interface diffusion by analyzing the distribution of radioactive material. He used autoradiographic methods after cutting successive thin layers from the surface under investigation.

The diffusion of lead in lead silicate was studied. It was found also, in silver sulphate, that the transport of material is due only to silver ions and not to sulphate ions. The use of tracer methods to measure transfer numbers was applied to calcium oxide and the diffusion of zinc was studied in zinc oxide and zinc-iron-spinel.

Mr. J. K. Lancaster, presenting a paper for *Mr. M. Kerridge* (Aldermaston, England) described investigations of wear of metals and metallic transfer under pressure and under friction. The amount of metal transferred under controlled friction depended greatly on cleanliness, finish, and hardness of surfaces. Autoradiograph records related to frictional force showed how increased friction and metallic transfer followed the breakdown of a protective film of fatty acid.

Mr. J. C. E. Button (Chester, England) has measured the wear of wire-drawing dies by inspection of drawn copper wire after activation of the dies. The dies were of tungsten carbide in a cobalt matrix. Autoradiography of lengths of drawn wire showed that cobalt is transferred evenly, while the tungsten carbide is removed in discrete particles. Measurements were also made with coils of drawn wire surrounding a Geiger counter. Preliminary results indicated that die-wear fluctuated during the tests, which represented only a small fraction of the natural life of the die.

Mr. E. W. Voice (London) has used radon to determine gas-transit time in a blast furnace. Radon was injected into the input air stream in a short burst by means of explosives. Samples of the outlet gas were taken at intervals from two positions at the top of the burden and tested for radioactivity. Mr. Voice also described the use of small pellets of Co^{60}, built into the refractory lining of a blast furnace. The radioactivity from the pellets can be detected from outside as an index of refractory wear, and can be detected in the metal output of the furnace if released.

Mr. C. B. Alcock (London), using S^{35} as a tracer, has investigated equilibrium conditions between $FeS(\beta) + H_2$ and $Fe + H_2S$ at temperatures from 450 to 980° C. The partial pressure of H_2S in the circulating gases was measured by using a thin-window beta counter. Measurements of relative vapor pressures of H_2 and H_2S were made as a function of gas-flow rate. These gave

quantitative data as to the thermal segregation errors involved in experiments at low gas-flow rates and enabled earlier results for the equilibrium to be reconciled with calorimetric data.

Determination of Silver in Photographic Images

The use of I^{131} to determine the silver in developed photographic images was described by *Dr. G. W. Stevens* (Harrow, England) in collaboration with *Mr. A. E. Ballard* and *Dr. C. W. Zuelke* (Rochester, New York). Two methods were described by which the silver was converted in situ to silver iodide labeled with I^{131}. The image was then scanned with an end-window beta counter, either by punching samples from a film, or by using a suitable aperture in a mask to limit the area of inspection. Very good agreement was found with measurements by the classical potentiometric method, and a statistical accuracy of 1.5% was obtained. The image can be recovered after the measurement by treatment in a sodium sulfide bath.

Industrial Radiography

In a session on gamma radiography with radioisotopes, industrial applications to special and routine examinations were described. The use of Co^{60}, Ta^{182}, and Ir^{192} to examine steel castings and welds was described by a number of speakers experienced in the techniques.

The radioactive source method has been found preferable to the X-ray method in adaptability and maneuverability. It also has considerable economic advantages in the radiography of large thicknesses of steel for which the installation costs of high-energy X-ray equipment would be prohibitive. In one firm, gamma-ray exposures of iron and steel castings are made continuously throughout the day and night, and five or six castings are usually examined at once.

Mr. B. Schuil (Overschie, Netherlands) described applications of Ir^{192} sources in conjunction with Geiger counters to measure the wall thickness of boilers, drums, and drill holes.

Mr. W. S. Eastwood (Harwell) outlined the problems attending the irradiation, measurement, storage and supply of large quantities of Co^{60}, Ta^{182}, and Ir^{192} sources intended for gamma radiography.

Handling procedures and the use of specially designed source containers were discussed.

Nondestructive Testing

In two papers, *Mr. K. Fearnside* (Aldermaston) and *Mr. G. Syke* (Dartford, England) described commercial equipment using the absorption and scattering of beta particles and gamma rays for routine measurements of thickness.

Mr. Fearnside described a beta-transmission gage employing balanced sources and ionization chambers for measuring thickness fluctuations from a standard value. The use of a back-scattering-type beta gage to measure the thickness of coatings and liquid films on a metal backing was described.

A gage employing the scattering of Co^{60} gamma rays for thickness measurements up to 0.2 inch of hot-rolled steel strip has been developed.

Rejection of empty or incompletely filled sealed packages is facilitated by the use of a monitor which measures the absorption of beta particles in the packet as registered by a halogen-quenched Geiger counter.

Mr. G. Syke described beta-transmission gages employing a single ion chamber and source, with a provision for back-biassing the recorder to register a standard thickness. Balanced units are used when a thickness of coating, such as typewriter carbon on paper, is to be measured. The apparent absorption of beta particles in foils has been

increased by using collimators consisting of parallel plates attached to both the source and the detector. A substantially parallel beam of beta particles is thus incident on the foil to be measured and those particles which are scattered by the foil through more than a small critical angle are unable to enter the detector. It is claimed that 1% variations in thicknesses of from 1 to 2 mg/cm² can be measured by this means.

A collimated beam of gamma rays has been used in conjunction with a counter and rate meter for rapid routine tests of internal positioning in all-metal assemblies. The method has advantages in speed over radiography and fluoroscopy when large numbers of standard units are inspected.

Elimination of Static Electric Charges

Dr. P. S. H. Henry (Manchester, England) gave an account of the occurrence of static electric charges in the textile industry. He noted that charges are developed on threads during the warping process at too high a rate to be eliminated by radioactive static eliminators of manageable activity. However, the phenomenon of "fog marking," caused by the attraction of dust particles to charged warps left in a loom overnight, can be prevented by the use of low-activity sources. Eliminators using 1 or 2 mc Tl^{204} have been found adequate. Compared to beta emitters, alpha emitters are, in general, more expensive and likely to create more health hazards. Alphas offer the advantage of higher local ionization efficiency, but for this application such efficiency is not necessary.

Radioactivation Analyses

The conference ended with two papers on the assay of impurities by radioactivation analysis.

Mr. A. A. Smales (Harwell) reviewed the subject and showed that when substances of high neutron activation cross section are to be estimated, chemical separation may not be necessary. Usually, however, the required element is separated after addition of an inactive carrier element. Isotopes present in high enough concentrations are identified by absorption and half-life measurements. The limiting sensitivity of the method depends on the neutron flux available.

By irradiation in the Harwell pile, arsenic has been determined in quantities down to 10^{-10} gm. In biological material such as hair, nails, urine and blood, arsenic is detectable in proportions from 0.1 to 1 ppm, using very small samples (e.g., 50 microliters of blood). The natural arsenic content can thus be determined. Arsenic in germanium oxide can now be detected to better than 0.01 ppm.

Other examples of determinations by this method, all without chemical separations, included Rb in K salts, HfO_2 in ZrO_2, and Ir in Rh. Uranium can be detected to 1 ppm using a few hundred mg of sample by estimating the barium produced in fission products.

In a previous paper, *Dr. P. R. Lewis* (Cambridge) had described the use of radioactivation to determine sodium and potassium in nerves. Analyses had a standard error of about 2% in samples containing quantities of less than 1 μg of sodium and a few μg of potassium.

Professor G. Chaudron (Vitry-sur-Seine, France) reported the assay of impurities in very pure aluminum by radioactivation. Quantities of copper, sodium, and rare earths were determined. For instance, 0.1 ppm of copper was estimated to $\pm 2\%$ and 1 ppm of sodium to $\pm 5\%$. Blank experiments were made to avoid the prodution of Na^{24} by the (n,α) reaction on the aluminum. Copper was separated as a sulfide, the rare earths as hydroxides. Analysis of the decay curves enabled each impurity to be identified.

Calculation of yields in

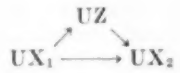
Multiple Neutron Capture Processes

In conversion reactors for the production of fissionable material and in breeders, neutron economy is vital. Presented here is a general formalism for evaluating effects of neutron capture by the initial nucleus and its products. Data just declassified are used in an example of the method

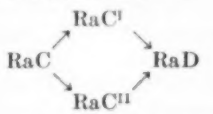
By R. F. VAN WYE* and J. G. BECKERLEY†

BOMBARDMENT OF NUCLEI with pile neutrons in some instances involves the capture of neutrons not only by the starting nucleus but also by its immediate products. This effect may be appreciable where the pile flux is high. Such multiple neutron capture processes and the accompanying transformations (alpha and beta decay or fission) might, for example, influence the conversion of fertile material to fissionable material both by producing nuclei isotopic with the fissionable nuclei, thus diluting the fissionable material, and by producing "poisoning" nuclei.

It is the purpose of this paper to present a general formalism for handling the transformation sequences resulting from such processes. In the conventional transformation processes exemplified by the natural radioactive series or the fission-product chains, we are dealing with linear sequences with occasional branching, usually no more complicated than a triangular sequence such as



or a square sequence such as



These chains can be handled by matrix representation.‡

The present situation cannot be represented by a linear sequence with occasional branching, but must be represented by a network. A specific instance of the type of reaction under

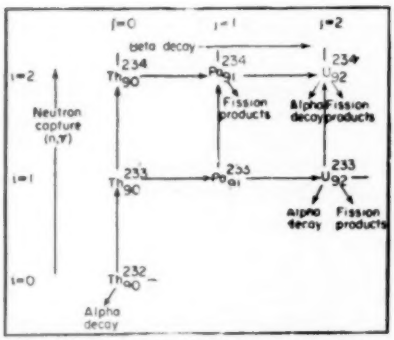


FIG. 1. Scheme of reactions for the irradiation of thorium by thermal neutrons in a pile

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‡ W. H. Watson, A method for computing transformations in radioactive series, *Can. J. Research* **25A**, 252 (1947); H. Jeffries, B. S. Jeffries, "Methods of Mathematical Physics" (University Press, Cambridge, 1950)

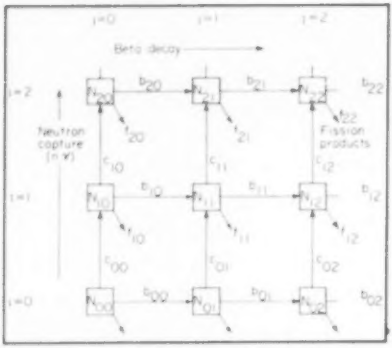

FIG. 2. General scheme for neutron capture, beta decay, and fission for thermal-neutron irradiation of materials in a pile. The vertical arrows indicate neutron capture, the horizontal arrows beta decay, and the diagonal arrows fission

consideration here is shown in Fig. 1. It is assumed that the neutrons are slow and that only (n,γ) transformations are important, e.g., $(n, 2n)$ reactions are not considered. Some of the isotopes that are produced decay by alpha emission, some by beta emission and some by fission. The possibility of an alpha decay followed by neutron capture can be neglected in most cases of interest to reactor engineers. This is because the shortest alpha-decay half-life is usually large compared to the longest beta-decay half-life.

The generalization of the network of Fig. 1 is shown in Fig. 2. Each N_{ij} indicates an isotope with i representing the change in mass number and j the change in atomic number. Neutron capture is indicated by a vertical arrow, beta decay by a horizontal arrow. Since fission produces nuclei whose mass and atomic numbers are outside the range of the grid, diagonal arrows indicate fission as removing nuclei from the grid. (Alteration of the neutron flux by the presence of fission products, as well as all other nuclei, is neglected.) In Fig. 2 the b's, c's, and f's are the probabilities per second for beta decay, neutron capture, and fission, respectively. The c's and f's are the flux times the capture and fission cross sections, respectively, while the b's are the usual beta disintegration constants.

Consider any (i,j) nucleus; see Fig. 3. The rate of change of N_{ij} is the sum of production rates (incoming arrows) less the sum of decay, capture, and fission rates (outgoing arrows). Thus

$$dN_{ij}/dt = (b_{i,j-1}N_{i,j-1} + c_{i-1,j}N_{i-1,j}) - k_{ij}N_{ij} \quad (1)$$

where

$$k_{ij} = b_{ij} + c_{ij} + f_{ij}$$

and

$$b_{ij} = \lambda_{ij} = \text{beta disintegration constant}$$
$$c_{ij} = \phi\,\sigma_c(ij)$$
$$f_{ij} = \phi\,\sigma_f(ij)$$

in which ϕ is the thermal-neutron flux, $\sigma_c(ij)$ is the thermal-neutron capture cross section, and $\sigma_f(ij)$ is the thermal-neutron fission cross section. The values of i and j are never less than zero.

The quantity of any component isotope, N_{ij}, existing at any time in the nuclear processes represented by the grid in Fig. 2 is given in the solution of Eq. 1. The activity of the component is given by the product of N_{ij} and the proper function of the disintegration constant and fission rate. The solution of Eq. 1 is

$$N_{ij} = \sum_{m=0}^{i}\sum_{n=0}^{j} A_{ij}^{mn}\, e^{-k_{mn}t} \quad (2)$$

The coefficients A_{ij}^{mn} are functions of b_{ij}, c_{ij}, and k_{ij}, and may be determined from a three-term recursion formula. This recursion formula is found by substituting N_{ij} from Eq. 2 into Eq. 1, and equating coefficients of $e^{-k_{mn}t}$. The result is

$$A_{ij}^{mn} = \frac{b_{i,j-1}}{k_{ij} - k_{mn}} A_{i,j-1}^{mn} + \frac{c_{i-1,j}}{k_{ij} - k_{mn}} A_{i-1,j}^{mn} \quad (3)$$

The second term of this equation

TABLE 1 — Values of the Expansion Coefficient, $A_{ij}{}^{mn}$, for Values of m and n Ranging from 0 to 2

$$A_{00}{}^{00} = N_{00}{}^{0}$$
$$A_{01}{}^{00} = b_{00}N_{00}{}^{0}/(k_{01} - k_{00})$$
$$A_{01}{}^{01} = N_{01}{}^{0} - \frac{b_{00}N_{00}{}^{0}}{k_{10} - k_{00}}$$
$$A_{10}{}^{00} = c_{00}N_{00}{}^{0}/(k_{10} - k_{00})$$
$$A_{10}{}^{10} = N_{10}{}^{0} - \frac{c_{00}N_{00}{}^{0}}{k_{10} - k_{00}}$$
$$A_{20}{}^{00} = c_{10}A_{10}{}^{00}/(k_{20} - k_{00})$$
$$A_{20}{}^{10} = c_{10}A_{10}{}^{10}/(k_{20} - k_{10})$$
$$A_{20}{}^{20} = N_{20}{}^{0} - \frac{c_{10}A_{10}{}^{10}}{k_{20} - k_{10}} - \frac{c_{10}A_{10}{}^{00}}{k_{20} - k_{00}}$$
$$= N_{20}{}^{0} - A_{20}{}^{10} - A_{20}{}^{00}$$
$$A_{02}{}^{00} = b_{01}A_{01}{}^{00}/(k_{02} - k_{00})$$
$$A_{02}{}^{01} = b_{01}A_{01}{}^{01}/(k_{02} - k_{01})$$
$$A_{02}{}^{02} = N_{02}{}^{0} - \frac{b_{01}A_{01}{}^{01}}{k_{02} - k_{01}} - \frac{b_{01}A_{01}{}^{00}}{k_{02} - k_{00}}$$
$$= N_{02}{}^{0} - A_{02}{}^{01} - A_{02}{}^{00}$$
$$A_{11}{}^{00} = \frac{b_{10}A_{10}{}^{00}}{k_{11} - k_{00}} + \frac{c_{01}A_{01}{}^{00}}{k_{11} - k_{00}}$$
$$A_{11}{}^{01} = c_{01}A_{01}{}^{01}/(k_{11} - k_{01})$$
$$A_{11}{}^{10} = b_{10}A_{10}{}^{10}/(k_{11} - k_{10})$$
$$A_{11}{}^{11} = N_{11}{}^{0} - \frac{c_{01}A_{01}{}^{01}}{k_{11} - k_{01}} - \frac{c_{01}A_{01}{}^{00}}{k_{11} - k_{00}} - \frac{b_{10}A_{10}{}^{10}}{k_{11} - k_{10}} - \frac{b_{10}A_{10}{}^{00}}{k_{11} - k_{00}}$$
$$= N_{11}{}^{0} - A_{11}{}^{01} - A_{11}{}^{10} - A_{11}{}^{00}$$
$$A_{12}{}^{00} = \frac{b_{11}A_{11}{}^{00}}{k_{12} - k_{00}} + \frac{c_{02}A_{02}{}^{00}}{k_{12} - k_{00}}$$
$$A_{12}{}^{01} = \frac{b_{11}A_{11}{}^{01}}{k_{12} - k_{01}} + \frac{c_{02}A_{02}{}^{01}}{k_{12} - k_{01}}$$
$$A_{12}{}^{02} = c_{02}A_{02}{}^{02}/(k_{12} - k_{02})$$
$$A_{12}{}^{10} = b_{11}A_{11}{}^{10}/(k_{12} - k_{10})$$
$$A_{12}{}^{11} = b_{11}A_{11}{}^{11}/(k_{12} - k_{11})$$
$$A_{12}{}^{12} = N_{12}{}^{0} - \frac{b_{11}A_{11}{}^{00}}{k_{12} - k_{00}} - \frac{b_{11}A_{11}{}^{01}}{k_{12} - k_{01}} - \frac{b_{11}A_{11}{}^{10}}{k_{12} - k_{10}} - \frac{b_{11}A_{11}{}^{11}}{k_{12} - k_{11}} - \frac{c_{02}A_{02}{}^{00}}{k_{12} - k_{00}} - \frac{c_{02}A_{02}{}^{01}}{k_{12} - k_{01}} - \frac{c_{02}A_{02}{}^{02}}{k_{12} - k_{02}}$$
$$= N_{12}{}^{0} - A_{12}{}^{00} - A_{12}{}^{01} - A_{12}{}^{02} - A_{12}{}^{10} - A_{12}{}^{11}$$
$$A_{21}{}^{00} = \frac{b_{20}A_{20}{}^{00}}{k_{21} - k_{00}} + \frac{c_{11}A_{11}{}^{00}}{k_{21} - k_{00}}$$
$$A_{21}{}^{01} = c_{11}A_{11}{}^{01}/(k_{21} - k_{01})$$
$$A_{21}{}^{11} = c_{11}A_{11}{}^{11}/(k_{21} - k_{11})$$
$$A_{21}{}^{10} = \frac{b_{20}A_{20}{}^{10}}{k_{21} - k_{10}} + \frac{c_{11}A_{11}{}^{10}}{k_{21} - k_{10}}$$
$$A_{21}{}^{20} = b_{20}A_{20}{}^{20}/(k_{21} - k_{20})$$
$$A_{21}{}^{21} = N_{21}{}^{0} - \frac{b_{20}A_{20}{}^{00}}{k_{21} - k_{00}} - \frac{b_{20}A_{20}{}^{10}}{k_{21} - k_{10}} - \frac{b_{20}A_{20}{}^{20}}{k_{21} - k_{20}} - \frac{c_{11}A_{11}{}^{00}}{k_{21} - k_{00}} - \frac{c_{11}A_{11}{}^{01}}{k_{21} - k_{01}} - \frac{c_{11}A_{11}{}^{10}}{k_{21} - k_{10}} - \frac{c_{11}A_{11}{}^{11}}{k_{21} - k_{11}}$$
$$= N_{21}{}^{0} - A_{21}{}^{00} - A_{21}{}^{01} - A_{21}{}^{11} - A_{21}{}^{10} - A_{21}{}^{20}$$
$$A_{22}{}^{00} = \frac{b_{21}A_{21}{}^{00}}{k_{22} - k_{00}} + \frac{c_{12}A_{12}{}^{00}}{k_{22} - k_{00}}$$
$$A_{22}{}^{01} = \frac{b_{21}A_{21}{}^{01}}{k_{22} - k_{01}} + \frac{c_{12}A_{12}{}^{01}}{k_{22} - k_{01}}$$
$$A_{22}{}^{02} = c_{12}A_{12}{}^{02}/(k_{22} - k_{02})$$
$$A_{22}{}^{10} = \frac{b_{21}A_{21}{}^{10}}{k_{22} - k_{10}} + \frac{c_{12}A_{12}{}^{10}}{k_{22} - k_{10}}$$
$$A_{22}{}^{20} = b_{21}A_{21}{}^{20}/(k_{22} - k_{20})$$
$$A_{22}{}^{11} = \frac{b_{21}A_{21}{}^{11}}{k_{22} - k_{11}} + \frac{c_{12}A_{12}{}^{11}}{k_{22} - k_{11}}$$
$$A_{22}{}^{21} = b_{21}A_{21}{}^{21}/(k_{22} - k_{21})$$
$$A_{22}{}^{12} = c_{12}A_{12}{}^{12}/(k_{22} - k_{12})$$
$$A_{22}{}^{22} = N_{22}{}^{0} - \frac{b_{21}A_{21}{}^{21}}{k_{22} - k_{21}} - \frac{b_{21}A_{21}{}^{20}}{k_{22} - k_{20}} - \frac{b_{21}A_{21}{}^{11}}{k_{22} - k_{11}} - \frac{b_{21}A_{21}{}^{10}}{k_{22} - k_{10}} - \frac{b_{21}A_{21}{}^{01}}{k_{22} - k_{01}} - \frac{b_{21}A_{21}{}^{00}}{k_{22} - k_{00}} - \frac{c_{12}A_{12}{}^{12}}{k_{22} - k_{12}} - \frac{c_{12}A_{12}{}^{11}}{k_{22} - k_{11}} - \frac{c_{12}A_{12}{}^{02}}{k_{22} - k_{02}} - \frac{c_{12}A_{12}{}^{10}}{k_{22} - k_{10}} - \frac{c_{12}A_{12}{}^{01}}{k_{22} - k_{01}} - \frac{c_{12}A_{12}{}^{00}}{k_{22} - k_{00}}$$
$$= N_{22}{}^{0} - A_{22}{}^{00} - A_{22}{}^{01} - A_{22}{}^{02} - A_{22}{}^{10} - A_{22}{}^{20} - A_{22}{}^{21} - A_{22}{}^{12} - A_{22}{}^{11}$$

TABLE 2

Values of the Disintegration Constants and Cross Sections

Isotope	i	j	*Beta disintegration constant (fractional disintegrations per sec)*	*Thermal-neutron capture cross section, σ_c* (barns)*	*Thermal-neutron fission cross section, σ_f* (barns)*
Th^{232}	0	0	0	6.5	$\leq 2 \times 10^{-4}$
Th^{233}	1	0	4.92×10^{-4}	1,500	~ 0
Pa^{233}	1	1	2.92×10^{-7}	37	< 0.1
Th^{234}	2	0	3.27×10^{-7}	~ 0	~ 0
Pa^{234}	2	1	1.01×10^{-2}	~ 0	1,000†

* These values were obtained from various AEC sources and should not be considered the most accurate values; they are used here for illustrative purposes.

† This assumes the natural mixtures of the isomers UZ and UX_2; this may be invalid for that part of the Pa^{234} formed from neutron capture by Pa^{233}.

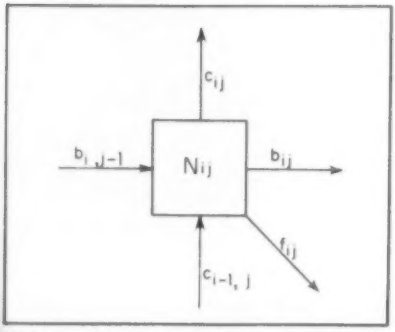

FIG. 3. Growth and decay processes for each N_{ij}

vanishes for $m = i$, and the first term vanishes for $n = j$, since Eq. 2 limits m and n to $m \leq i$ and $n \leq j$.

This relation is not valid for $(m,n) = (i,j)$. The coefficient A_{ij}^{ij} is determined by the conditions at $t = 0$, the initial conditions. Setting $t = 0$ in Eq. 2 gives

$$A_{ij}^{ij} = N_{ij}^0 - \sum_{mn}' A_{ij}^{mn} \qquad (4)$$

in which N_{ij}^0 is the value of N_{ij} at $t = 0$. The prime on the summation sign means that the sum is taken over m from 0 to i, and n from 0 to j, excluding the values $(m,n) = (i,j)$. Thus Eqs. 3 and 4 determine all the expansion coefficients in the solution of Eq. 2 for N_{ij}. Expansions from $n = 0$ to $n = 2$ and $m = 0$ to $m = 2$ are given in Table 1.

The expanded equations for N_{ij} and A_{ij} show how production of each component N_{ij} depends upon the preceding N_{ij} and the transformation probabilities expressed in the coefficients A_{ij}. For example, expand N_{20}, giving

$$N_{20} = \frac{c_{10}}{k_{20} - k_{00}} \frac{c_{00} N_{00}^0}{k_{10} - k_{00}} e^{-k_{00}t} + \frac{c_{10}}{k_{20} - k_{10}} \left(N_{10}^0 - \frac{c_{00} N_{00}^0}{k_{10} - k_{00}} \right) e^{-k_{10}t} + \left[N_{20}^0 - \frac{c_{10}}{k_{20} - k_{10}} \left(N_{10}^0 - \frac{c_{00} N_{00}^0}{k_{10} - k_{00}} \right) - \frac{c_{10}}{k_{20} - k_{00}} \frac{c_{00} N_{00}^0}{k_{10} - k_{00}} \right] e^{-k_{20}t}$$

This equation shows N_{20} as a function that depends on the exponential terms and the transformation coefficients operating on N_{00}^0, N_{10}^0, and N_{20}^0. Thus it shows that the quantity of N_{20} present depends on the initial quantity of each component isotope and the transformation processes in which it is involved.

The combination of the c and k terms in

$$\frac{c_{10}}{k_{20} - k_{00}} \frac{c_{00}}{k_{10} - k_{00}}$$

indicates the interaction of the c, b, and

TABLE 3

Quantities of Some Isotopes Produced When Thorium is Bombarded by Thermal Neutrons

Isotope	i	j	Time for equilibrium in flux of 10^{13} $n/cm^2/sec$ (sec)		Grams produced per gram of thorium in 10^7 sec		
					10^{12} $n/cm^2/sec$	10^{13} $n/cm^2/sec$	10^{14} $n/cm^2/sec$
Th^{233}	1	0	10^4	(3 hr)	10^{-8}	10^{-7}	10^{-6}
Th^{234}	2	0	10^7	(4 mo)	6×10^{-11}	6×10^{-9}	6×10^{-7}
Pa^{233}	1	1	10^7	(4 mo)	2×10^{-6}	2×10^{-4}	2×10^{-2}
Pa^{234}	2	1	10^7	(4 mo)	8×10^{-14}	8×10^{-12}	8×10^{-10}

f transformation probabilities. Further relationships and dependencies become apparent by examination of other coefficients and equations. The expanded equation for N_{22} would indicate the build up of N_{22} through several paths such as $N_{00}N_{10}N_{11}N_{12}N_{22}$, or $N_{00}N_{10}N_{20}N_{21}N_{22}$, or $N_{00}N_{10}N_{11}N_{21}N_{22}$.

Example

As an example, the quantities of Th^{233}, Th^{234}, Pa^{233}, and Pa^{234}, formed when thermal neutrons bombard Th^{232} in a reactor, can be calculated. It shall be assumed that at time $t = 0$ there is 1 gm of Th^{232} present.

Reference to Figs. 1 and 2 shows that Th^{233} is nucleus N_{10}, Th^{234} is N_{20}, Pa^{233} is N_{11}, Pa^{234} is N_{21}, and Th^{232} is N_{00}. Expansion of Eq. 2 for the appropriate values of i and j gives the equations for calculating the quantities of the isotopes present at time t. For the case $i = 2$ and $j = 1$

$$N_{21} = A_{21}{}^{00}e^{-k_{00}t} + A_{21}{}^{01}e^{-k_{01}t} + A_{21}{}^{10}e^{-k_{10}t} + A_{21}{}^{11}e^{-k_{11}t} + A_{21}{}^{20}e^{-k_{20}t} + A_{21}{}^{21}e^{-k_{21}t}$$

For the initial conditions of this example, all $N_{ij}{}^0 = 0$, except $N_{00}{}^0$. The A's as functions of the beta disintegration constants, b_{ij}, and the products of the flux and the cross sections, c_{ij} and f_{ij}, are given in Table 1. The disintegration constants and the thermal-neutron capture and fission cross sections are given in Table 2. Since Th^{233} has a long half-life for alpha emission, compared to the half-lives for the other processes involved, the alpha disintegrations are neglected.

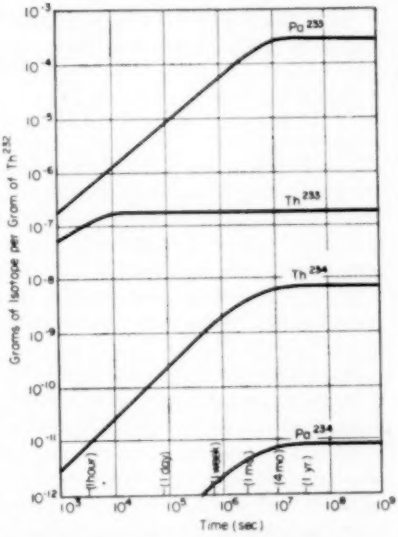

FIG. 4. Quantities of isotopes produced when thorium is transformed under bombardment by 10^{13} thermal neutrons per cm^2 per sec. Coordinates are on logarithmic scales

The calculated quantities of Th^{233}, Th^{234}, Pa^{233}, and Pa^{234} for fluxes of 10^{12}, 10^{13}, and 10^{14} thermal neutrons per cm^2 per sec, at $t = 10^7$ sec (about 4 months), when 1 gm of thorium is the starting material, are given in Table 3. The time for equilibrium* in a flux of 10^{13} $n/cm^2/sec$ is also given. Figure 4 shows the build up of these isotopes with time. —END

* Equilibrium occurs when the rate of formation of a particular isotope equals its rate of transformation into other isotopes, as shown in Fig. 3.

Relative Sensitivities of Windowless and End-Window Counters

Self-absorption in low-energy beta emitters limits sensitivity. This effect was studied and a comparison of counters was made with representative radioisotopes. Absorption data for 0–3 mg/cm² are presented for C^{14}, S^{35}, and Ca^{45}

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WINDOWLESS or internal counters operating in both the Geiger-Müller and proportional regions are now generally available. It seemed desirable, therefore, to compare the sensitivity of this type of instrument with that of the conventional thin mica end-window counter.

In general, the lower the beta energy being measured, the greater will be the advantage and consequently the usefulness of the internal counter. In the routine measurement of low-energy beta emitters, sensitivity is limited and a most important correction factor is caused by self-absorption in the sample being measured. It was necessary in a comparison of this type, therefore, to include a consideration of the effect of the type of counter upon the self-absorption characteristics.

Apparatus and Procedures

Three instruments were compared in this study:

1. Thin mica end-window counter. An Amperex model 100NB counter with a 3.5-mg/cm² mica window was used. This counter is filled with neon plus a halogen mixture, and operates at 700 volts with a plateau of approximately 200 volts. It has a stainless-steel cathode and is 1½ in. long, with a $1\frac{3}{16}$-in. outside diameter and a $\frac{3}{32}$-in. wall. This counter was used in a standard Lucite mount enclosed in a 2-in. lead shield. Samples were located 2 cm from the counter window. A conventional scaling unit with a scale of 64 was employed.

2. Windowless G-M flow counter. A Tracerlab model SC-16 windowless flow counter with end-on construction that gives nearly 50% geometry was used. Using a gas mixture of 99% helium and 1% isobutane, this counter operates in the Geiger region at approximately 1,300 volts. A 1-in. sample is accommodated in the counting chamber that is shielded by $1\frac{3}{8}$ in. of lead, and a three-position platform allows preflushing of the sample. The output of this counter was fed to a Neher-Pickering quench circuit and then to a conventional scaling unit with a scale of 256.

3. Windowless proportional counter. A Nuclear Measurements Corp. model PC-1 windowless proportional counter, designated as a restricted atmosphere counter, was used. The sample, which can be as large as 2¼ in. in diameter and ½ in. thick, is placed internally with a geometry approaching 50%. The counting chamber is not shielded. Employing a gas mixture of 90% argon, 10% methane, the counter operates at

about 1,700 volts. The voltage supplies, amplifying circuits and scale-of-512 are built in a single unit.

Dead-time measurements were made with Ca^{45} using the divided source method described by Rainwater and Wu (*1*). All activity measurements were subject to the usual corrections for background, dead-time, decay, and statistical deviation.

All measurements, except where otherwise noted, were made in 1-in. diameter stainless-steel cups. For the sensitivity comparisons a high-specific-activity solution of each isotope was evaporated in the cup to produce a sample of essentially zero mass. In the case of C^{14}, however, a small amount of powdered $BaC^{14}O_3$ was suspended in a Na_2SO_4 solution, and after standing overnight the solution was centrifuged and a small amount of the supernate was evaporated in the stainless-steel cup. For the UX_2 measurements a reference source was used in which the activity was spread over a brass disk of 23-mm diameter radiating area $\times$ 5/16 in. thick, and covered with aluminum foil of 150 mg/cm^2.

Calcium oxalate samples were used for the self-absorption determinations. These samples were prepared as follows: High-specific-activity $Ca^{45}Cl_2$, as obtained from Oak Ridge National Laboratory, was diluted with water to give an activity of approximately 4,000 cpm/ml as determined with an end-window G-M tube. A $CaCl_2$ solution containing 2.00 mg/ml of calcium was prepared by suspension of 5.0344 gm of chemically pure $CaCO_3$ in water, addition of 70 ml of 2*N* HCl and dilution to 1,000 ml. Varying amounts of the $CaCl_2$ solution were placed in 50-ml conical-bottom centrifuge tubes, and 1 ml of the active calcium solution was added to each. The volumes were adjusted to 25 ml when necessary by the addition of 0.01*N* HCl; 2 ml of 25% acetic acid and one drop of methyl red indicator were added. After the addition of 3 ml of saturated ammonium oxalate, the *p*H was slowly adjusted to the orange end point of methyl red (*p*H 4.0–5.0) with 3*N* NH_4OH. Acetic acid was added until the solutions were faintly pink, and the solutions were allowed to stand overnight.

The samples were then centrifuged and washed with two 10-ml portions of 2*N* NH_4OH saturated with CaC_2O_4. The precipitates were slurried in water and quantitatively transferred to stainless-steel cups in which plastic chimneys were fitted. The precipitates were centrifuged and the supernate syphoned off. The plastic tubes were removed and the samples dried under a heat lamp. This general method for the precipitation of CaC_2O_4 and collection of the precipitate has been described in detail by Comar *et al.* (*2*).

The $CaC_2O_4 \cdot H_2O$ precipitates were of uniform appearance except for some of the thin ones, which were not evenly distributed. Four samples were prepared at each thickness and counted. The weightless (<0.1 mg/cm^2) samples

OVER-ALL EFFICIENCY will depend on very many factors including such things as the size of the sample that a counter can accept, the background of the instrument, the ratio of the activity to be measured to the mass of the sample in which the activity is contained, and also to self-absorption. It would become very complicated to generalize in respect to all of these factors; therefore, this article presents the basic information which will enable the reader to apply this information to his own particular conditions.

TABLE 1
Relative Sensitivity Values for Weightless Samples*

Isotope	Energy (Mev)		Thin mica end-window G-M counter	Windowless G-M flow counter	Windowless proportional counter
	Beta	Gamma			
C^{14}	0.155		0.031	1.0	1.1
S^{35}	0.167		0.030	1.0	1.2
Ca^{45}	0.255		0.13	1.0	1.5
Co^{60}	0.31	1.17, 1.33	0.070	1.0	1.1
Ta^{182}	0.53	1.13, 1.22, 1.24	0.10	1.0	1.2
I^{131}	0.60, 0.32	0.364, 0.284, 0.080, 0.638	0.13	1.0	1.2
Cs^{134}	0.658, 0.090	0.602, 0.794, 1.35, 0.565	0.088	1.0	0.95
P^{32}	1.69		0.24	1.0	1.7
Sr^{90}-Y^{90}	0.537, 2.18		0.18	1.0	1.8
UX_2†	2.32		0.32	1.0	1.2
Fe^{55}	K		0.006	1.0	1.9
Average Background (cpm)			17	31	72
Deadtime (μsec)			236	150	1.28

* These values represent the relative counting rate for the same sample of each isotope.
† ¼ in. closer to sensitive volume than other sources.

were prepared by evaporating 1 ml of the active $Ca^{45}Cl_2$ solution.

Sensitivity Comparisons

It is apparent that a comparison of sensitivities, particularly when the G-M tube is involved, will be somewhat arbitrary, depending primarily upon the characteristics of the particular tubes and the effective distances between the samples and the sensitive volumes of the tubes. The values reported here are based upon routine conditions used in this laboratory, and, while they may not be accurately applicable elsewhere, they can be used as a basis for comparison.

In this work, a 3.5-mg/cm² end-window tube was employed rather than one with a thinner window because certain fundamental differences, particularly in the self-absorption curves, become more evident. The greater mechanical strength of the 3 to 4 mg/cm² tube, as compared with that of tubes with thinner windows, has prompted the use of the former whenever possible in this laboratory. The increase in sensitivity for radiations from Ca^{45}, S^{35}, C^{14}, P^{32}, Au^{198}, I^{131}, and Co^{60}, as the window thickness is decreased, can be estimated by using the absorption correction factors calculated by Gleason *et al.* (*3*). To adjust for differences in the distance between sample and window, both the air absorption and the geometry factor can be estimated.

Comparative sensitivities of the three counters for several isotopes are shown in Table 1. The values given represent the relative counting rates as found with the same radioisotope sample.

In making a critical comparison of sensitivities it is necessary to take into account the background of the counter (*4*). However, the background is not entirely an intrinsic quality of the counter and may be modified by the shielding; furthermore, the merit of one counter over another will depend upon the counting rates to be used. Therefore, the values have been expressed as

the actual observed comparative counting rates.

It is apparent from Table 1 that the internal counters were about 3 to 30 times more sensitive than the end-window counter, with the differences being more pronounced for the soft-beta emitters. In general the proportional counter showed counting rates about 1.2 times those of the windowless G-M counter.

Self-Absorption Considerations

In routine experimentation the preparation of all samples of zero thickness is virtually impossible. Also, the preparation of all samples at a standard thickness is inconvenient and impractical. A general discussion of procedures used for self-absorption corrections has been reported by several workers (*2*, *5–9*). The most widely used method for evaluation of the observed activity is to convert this value to the activity which would have been observed at some specified thickness.

The most commonly used thicknesses for reference are zero thickness or infinite thickness (maximum observed activity for increasing amounts of a sample of constant specific activity). In preparation of a set of standards of constant specific activity, the determination of the value of infinite thickness is difficult because the observed activity increases in the following manner

$$N/N_\infty = 1 - e^{-\mu T_c}$$

where N is the observed activity, N_∞ is the activity at infinite thickness, T_c is the sample thickness in mg/cm², and μ is the self-absorption coefficient in cm²/mg.

Figure 1 shows such a constant-specific-activity curve for S^{35}. The slope at thickness equal to zero, which is represented by the broken line N_0, represents the relationship between activity and weight of sample if no self-absorption occurred. Uncertainties in the determination of this slope are due

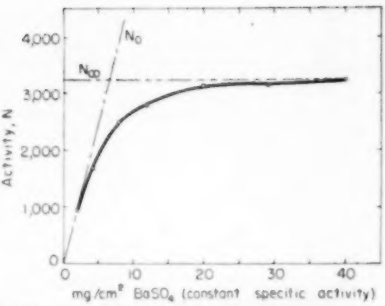

FIG. 1. Effect of self-absorption on measured activity of S^{35} in barium sulfate of constant specific activity

to self-scattering of the beta particles by the thin samples and to the difficulties in obtaining a uniform spread of the thin samples. Uncertainties in determination of activity at infinite thickness, N_∞, are due to the nature of the curve as it approaches zero slope, which may be complicated by the fact that the surface gets closer to the window as the sample thickness is increased.

The equation for self-absorption where N_0 is the true activity of the sample and T represents the sample thickness in mg/cm², there being a constant amount of sample activity, is

$$N/N_0 = (1 - e^{-\mu T})/\mu T$$

If a set of self-absorption standards is prepared using a constant amount of activity for each sample, the resultant self-absorption curve may be used to correct the observed activity to the activity which would be measured if the samples were weightless. In this method, the reference point (activity of a weightless sample) is well defined and not artificial.

Comar *et al.* (*2*) have used this method in the measurement of Ca^{45} (E_{max} = 0.260 Mev) in calcium oxalate precipitates counted with end-window G-M tubes. It was found that the ratio of the observed activity at a definite thickness to the activity of a weightless sample is reasonably independent of the window thickness in the

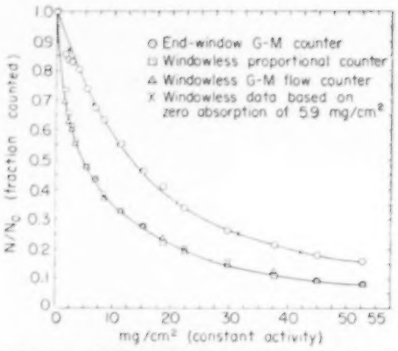
○ End-window G-M counter
□ Windowless proportional counter
△ Windowless G-M flow counter
x Windowless data based on zero absorption of 5.9 mg/cm²
N/N₀ (fraction counted)
mg/cm² (constant activity)

FIG. 2. Self-absorption curves for Ca^{45} in calcium oxalate as measured with end-window and windowless counters

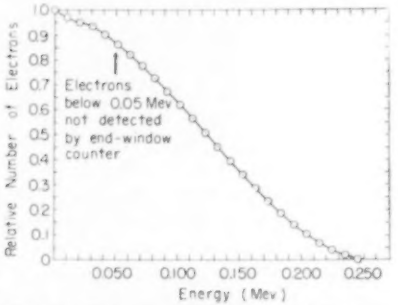

FIG. 3. Beta-particle energy distribution of Ca^{45}

range 1.8 to 3.5 mg/cm² and also is almost independent of the distance from the sample to the window of the counting tube as measured on the second to fifth shelves of a standard sample holder. The curve is established by measurement of samples containing a constant activity but varying mass, and calculations based on the observed activity of the weightless sample.

In use, a table is made of the N_0/N value for each sample thickness, and the observed counting rate is multiplied by the appropriate N_0/N to give the value which would be observed if the sample were weightless.

Self-absorption curves for Ca^{45}, measured as calcium oxalate in the three counters being compared, are given in Fig. 2. For the end-window G-M counter, the self-absorption coefficient was found to be 0.12 cm²/mg; this value is in agreement with the value of 0.11 reported by Comar *et al.* (*2*) and that of 0.09 reported by Glendenin and Solomon (*10*).

Sufficient information is not available for the other isotopes to calculate their self-absorption coefficients. Coefficients for the windowless counters are not presented because the self-absorption curve for these counters does not obey the same equation as for the end-window counter, and the computed coefficient would not be theoretically sound.

The curves for the two internal-type counters (Fig. 2) are essentially identical. In comparing the values obtained on the end-window counter with those obtained on the internal counters, it is noted that N/N_0, or the fraction counted, falls off more rapidly for the internal counters as the sample thickness is increased. This may be explained by referring to Fig. 3 which presents the beta-particle distribution for Ca^{45} (*11*). Since the air space and mica window was equivalent to about 5.9 mg/cm², it is apparent that electrons of less than 0.05 Mev would not reach the sensitive volume of the end-window counter. This means that the internal counters are measuring a greater proportion of the very-low-energy betas than is the window counter. Consequently, the observed attenuation of the activity by the sample itself will be greater when measured by the internal counter because of the increased proportion of low-energy electrons. If the values for the internal counters are calculated on the basis of an N_0 value of 5.9 mg/cm², the curve in Fig. 2 becomes identical with that obtained by the end-window measurements. This indicates that the window and air space attenuation is the primary cause of the difference between

the end-window and windowless self-absorption curves.

In consideration of the effect of self-absorption characteristics on the over-all sensitivity comparisons, it will be noted that there is the tendency to reduce the advantage of the internal counters. However, in the case of Ca^{45} there is still a considerable advantage of the windowless over the window counter. This is shown in Table 2.

Absolute Absorption

Figure 4 shows the absorption of C^{14}, S^{35} and Ca^{45} as measured in the windowless proportional counter. In each case samples of essentially zero mass were prepared by evaporation of high-specific-activity solutions onto copper planchets. The planchet was placed on a brass disk, and aluminum leaf calibrated for thickness by weighing was held in place over the sample by a ring which insured intimate contact between sample and absorber. In this way absolute absorption measurements were obtained for 0 to 3 mg/cm².

As expected, the curves for sulfur and carbon were coincident in this range. Rather surprising was the change in slope noted with Ca^{45}. To the authors' knowledge this change of slope has not been previously reported. Practically all of the previous data have been obtained with end-window counters in which there was always absorption due to air space and window thickness; thus the absorption values previously reported in the literature for the range of 0 to 3 mg/cm² have, as far as is known, been obtained by extrapolation.

* * *

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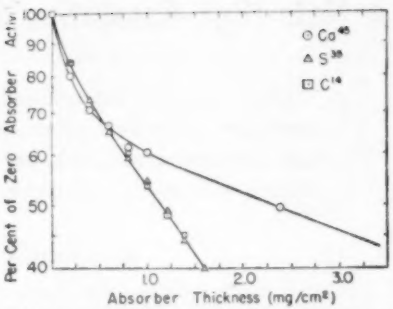

FIG. 4. Aluminum absorption curves of Ca^{45}, S^{35}, and C^{14} as measured with the windowless proportional counter

TABLE 2

Effect of Sample Thickness on Relative Sensitivity Values for Ca^{45}

	Sensitivity ratios	
Thickness (mg/cm²)	*Windowless G-M to end-window**	*Windowless proportional to end-window*
0	7.7	11.1
5 to 50†	5.0	7.0

* The counting rate of the windowless G-M counter divided by that of the end-window counter for the same sample of designated thickness.

† The values were independent of sample thickness in this range.

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A New High-Resolution System of Autoradiography*

Called "wet-process autoradiography," this unique system of radiation detection offers one-micron resolution with simple equipment. Single-cell studies and metal grain structure are examples of tracer research for which it is suited

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ONE-MICRON resolution in a radiation detection system has long been sought by research workers who use radioactive tracers. Resolution of this order can make possible studies of the life processes in single cells, detail analyses of the grain structure of metals, and many other investigations to which the tracer technique is well suited, but which have been hampered by inadequate radiation detection systems. A system that will provide such high resolution is called wet-process autoradiography. It requires only simple equipment and techniques.

In autoradiography, a radiation source is placed in contact with a piece of film or other radiation-sensitive material. The point at which the radiation leaves the surface of the source and enters the film can be identified after photographic development of the film. The distribution of radioisotopes in relation to microstructure, as determined by 1,000-diameter enlargement under the microscope, cannot, however, be determined by the simple autoradiographic technique just described. Difficulties arise due to film thickness, inert supporting layers or protective coatings on film, lack of intimate contact between the active surface and the film, low silver halide content in standard film emulsions, or specimen thickness. These and a number of other factors, together or separately, make it difficult to attain high resolution in autoradiography.

Considerable effort has gone into overcoming these difficulties, and good results have been obtained by a number of workers in this field. There is the gelatin-emulsion technique of Belanger and Leblond (*1*); the NTB stripping-film system of Boyd (*2*) and, using Ilford film, of Pelc (*3*); the specimen-on-film system of Evans (*4*); and a number of other variations of the placement of film or prepared emulsion with respect to the surface to be autoradiographed. Even these systems, however, have limitations that make achieving the desired resolution difficult.

In the present approach to the problem, a wet-process technique was evolved. A very thin and dense layer of sensitized silver bromide formed by chemical reaction directly on the surface to be studied is the basis of the technique. This layer, when formed, is about one micron thick and has no voids discernible under the microscope. The sensitivity is high, comparing favorably with the commerical films now available.

The sensitized layer, if processed properly, can be kept free of significant

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Wet-Process Autoradiography *has its roots in the old wet-collodion system of photography. Wet-collodion photography became obsolete toward the end of the 19th century with the advent of the dry-gelatin photographic plate. None of the modern concepts of the interaction of radiation with matter had been evolved at the time, and so the available literature on the wet-collodion process runs toward "cook book" formulas and collections of empirical data. Only one relatively scientific discussion and assembly of formulas exists, written by J. M. Eder* (5), *but this, too, by modern standards is wholly empirical.*

In the original wet-collodion process, a glass plate is coated with a solution of collodion in ether and alcohol in which a metal iodide such as cadmium iodide has been dissolved. When the collodion has set, but is not yet dry, the plate is transferred to a bath of silver nitrate in water. Silver iodide forms immediately and in two or three minutes the plate is ready for use. These plates must be used while still wet—hence the name of the process.

The films formed using the original formulas have many important basic faults—among them almost zero sensitivity to beta radiation and rapid loss of silver iodide to the silver nitrate solution. The original process proved to be quite unsuitable for autoradiography or use as a detector of ionizing particles. However, the physical operation of film formation on a surface proved so simple that the technique described below, using wet collodion as a base, was evolved.

fog or other artifacts. However, the chemicals used in forming the film are fairly active, and the problem of chemical interaction between the sensitized layer and the active substrate must be considered in the interpretation of any autoradiograph.

Plastic coatings, which are about one to two microns thick and which successfully inhibit direct chemical interaction, have been developed, but more work is needed on this problem. Where the protective layer is used, there is a corresponding loss in resolution.

Wet Autographic Technique

The formulas and directions for carrying out wet-process autoradiography are presented below. Special problems require variations in procedure to meet the conditions present.

The technique described herein is suitable for coating any surface, such as plastic or metal, to which a collodion layer will adhere without peeling when the surface is immersed in water. The preparation of such surfaces for tissue specimens in biological work or metallurgical specimens is discussed later.

The sensitized layer and developing solutions. The materials used for the layer, its application, and slide development are tabulated on the next page.

Physical conditions. The collodion and silver-nitrate solutions may be stored conveniently in screw-top, 70-cm³-capacity Coplin stain jars.* They are especially suitable for work with microscope slides. If a completely dark storage cabinet is not available, the silver-nitrate jar must be made light-tight. This is done by painting the glass with two or more coats of glossy black enamel. If possible, the outside surface of the jar should be sandblasted first. The covers from these jars usually have a metal-covered insert. The metal coating of the silver-nitrate jar cover should be removed

* Available from the Arthur H. Thomas Co., Philadelphia, Pa. (Catalog No. 9196-S).

MATERIALS USED IN WET-PROCESS AUTORADIOGRAPHY

A. **Collodion layer** (composition of 50 cm^3 of solution); kept at 1° C

(1) Collodion: preferably USP solution (cellulose nitrate will work satisfactorily; celluloid or flexible collodion containing camphor should be avoided). Amount: $\frac{5}{8}$ gm dry weight or $15\frac{3}{4}$ cm^3 of USP solution

(2) Alcohol: absolute; to bring solution to $\frac{5}{8}$ gm of cellulose nitrate in 50 cm^3

(3) Cadmium bromide: 0.75 gm

(4) Ammonium bromide: 0.15 gm

B. **Silver nitrate bath;** kept at 1° C

(1) Silver nitrate: 25 gm

(2) Distilled water: 250 cm^3

(3) Sulfuric acid: to produce pH = 2.5 (about 16 drops of 10% solution of CP sulfuric acid will produce this pH)

(A small amount of silver sulfate will be precipitated.)

C. **Developing solutions**

(1) Silver nitrate bath as in *B* above, but maintained at room temperature

(2) Developer (at room temperature) as follows:
Ferrous sulfate: 10 gm
Distilled water: 250 cm^3
Alcohol: 7.5 cm^3
Acetic acid, glacial; adjustable and critical (see text); typical value: 7.5 cm^3

(3) Fixer: hypo at room temperature

to prevent attack by the silver nitrate. The cover of the collodion jar need not be disturbed.

Both collodion and silver-nitrate solutions should be kept at about 1° C. This is important to preserve the collodion and to prevent fog in the final film.

A standard beverage cooler with a circulating water bath is a very convenient cold-box. A small Coca Cola cooler with a refrigerating unit and a water circulator has proved to be satisfactory for this work. A splash shield to cover the water circulator is helpful. These units maintain temperature to within one degree uniformly throughout the box.

Manipulation in applying layer. Operations are carried out under a standard darkroom safelight. Actually, the reduced illumination is needed only when the specimen goes into the silver nitrate, but since it is inconvenient to bother with lights in the middle of the processing, carrying out the total procedure under the safelight is generally more satisfactory.

It is assumed that a properly prepared microscope slide is to be processed. The slide is dipped into the chilled collodion solution and then withdrawn. The specimen on the slide should be well covered and preferably located in the center of the coated area. The slide is allowed to drain and dry for thirty seconds. During this time, it is held vertical, and the lower end may be touched to paper toweling or any absorbent surface to help remove the surplus solution.

After thirty seconds, the slide is placed in the black stain-jar containing the cold silver-nitrate solution. The silver-nitrate jar is then covered and returned to the refrigerator. The same is done with the collodion-solution jar.

Nothing further is done until the total exposure time has elapsed. The slide is then removed, again under a safelight, and developed.

Manipulation during development. Under a safelight, the specimen slide is transferred from the cold to the room-temperature silver-nitrate solution. One minute is allowed for the slide to warm up. The slide is then transferred to a "developer slip" holding about 10 to 15 cm^3 of the fresh ferrous-sulfate solution. Developer solution is used once and then discarded.

The developer slip is a small container made of flattened glass tubing large enough to hold just one microscope slide. It must be deep enough so that the total area wet with silver-nitrate solution is well immersed in the developer. Such a slip is shown in Fig. 1.

Development is allowed to proceed for 30 seconds. The slide is then transferred to the hypo bath, where it will clear in about 15 seconds. One-minute fixation in active hypo is completely adequate. The slide is then rinsed gently and may be dried under a soft air blast. Collodion does not absorb water, so that drying consists essentially of removing surface water.

Where the facilities are adequate and the technique is understood, the slide is ready to go under the microscope within 4 or 5 minutes after being removed from the cold silver-nitrate bath.

The completed slide will consist of the original test specimen, its protective layer, if present, and a very thin autoradiographic over-all coating.

Detailed Analysis of the Process

The process has been adjusted so that only chemicals and equipment readily available in a university or hospital laboratory are required.

Use of collodion. The collodion serves two important functions. First, it acts as a mechanical support for the silver bromide grains, which are the actual radiation detectors. Second, it provides a region in which, by solubility difference, silver bromide may be formed. Silver bromide, being extremely insoluble in water, remains in the collodion layer on formation rather than diffusing out into the silver-nitrate solution.

The ready availability and widespread use of collodion make it a desirable material to use. Also, it has many desirable physical properties. When alcohol is used as a solvent, the solutions used in this process flow freely at room temperature and even at 1° C. Because volatile solvents are used, the film sets up very quickly, bringing handling time to a minimum. Being hydrophobic, the film does not swell up in water and, therefore, has very good dimensional stability.

Actually, other film materials could be used. Experimental films have been made with Formvar and with polystyrene, but they have not proved as easy to handle as the collodion films.

Gelatin, being water soluble unless specially treated, is not a suitable material for this process because long water immersion is required.

The amounts of collodion or cellulose nitrate in the coating solution and the amount of dissolved bromide (about 1

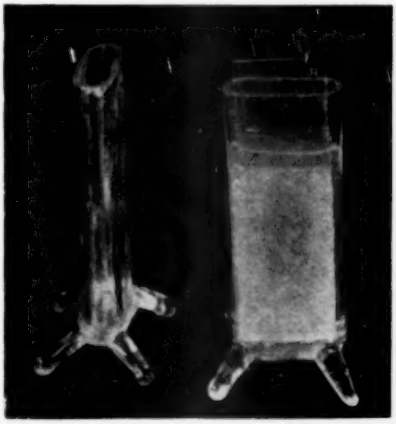

FIG. 1. Glass developer slip and a silver-bromide-coated tissue slide being developed in ferrous sulfate. Body of the developer slip is flattened glass tubing

gm in 50 cm³ of solution) have been adjusted so that the film will contain a monograin layer of silver bromide at the end of the process. More collodion will produce a thicker coat, and less collodion will lead to flaking and chipping of the film because it is incapable of holding in suspension the silver bromide formed in the film.

A proportional reduction in both bromide and collodion leads to a film that is satisfactory mechanically but has too much empty area because its silver bromide is less dense. Such films are useful where the grain structure of the film is of interest, but are not suitable for good autoradiography.

Silver-bromide emulsions have two important advantages over silver-iodide emulsions. First, the bromide films have good radiation sensitivity. Second, silver bromide does not dissolve in the silver-nitrate solutions. Films have been kept in the solution for as long as a week without loss in density.

Silver-bromide films, however, are quite susceptible to fog or deposition of silver in areas not affected by radiation. This tendency has been successfully suppressed by using the temperature cycle previously discussed and by controlling the *p*H of the silver-nitrate solution.

The use of mixed bromides, such as cadmium and ammonium bromides, arises from the effect of the cadmium and ammonium ions on the viscosity of the solution. The presence of cadmium ions tends to increase the viscosity of the solution, while ammonium ions tend to decrease it. This was observed by the early workers in collodion photography. By mixing the ions as indicated in the formulas given previously, the viscosity of the final mixture is essentially the same as that of the original collodion. Two essential properties of the metallic ion used are that it have a water-soluble nitrate so that no precipitate will form in addition to the silver halide, and that the metallic halide be soluble in alcohol.

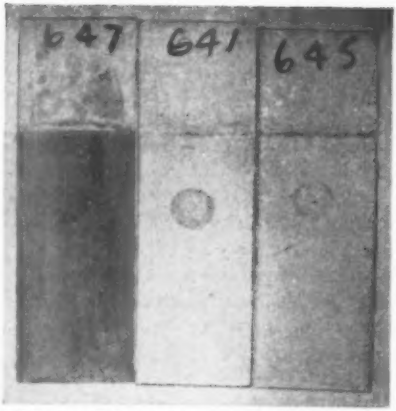

FIG. 2. Effect of temperature control on fog and sensitivity. Slide 647 was processed with all solutions at room temperature. Slide 645 was processed with all solutions at 4° C. Slide 641 was processed using the temperature cycle described in the text. All slides had equally active areas of radioactive scandium oxalate and all were exposed for the same length of time

Sensitization. In investigating the merits of this process, studies were made on the possibilities of increasing the sensitivity of the grains. Early attempts, using very small amounts of ammonium sulfide in the collodion solutions showed great promise, but these films were more easily fogged than those free of the sulfide.

This method of sensitization, based on the introduction of divalent impurities to form lattice ion vacancies and to create sensitivity specks, is no longer in use. It was found difficult to control and to duplicate results.

Quantitative tests, as reported here, indicate that each grain requires, on the average, 2 hits from I^{131} beta particles to render a grain developable. This sensitivity is adequate for most work and results in somewhat less susceptibility to fog and cosmic-ray tracks than more sensitive grains.

Further studies of sensitization, consisting of introducing sulfide impurities in a more easily controlled form and affecting crystal lattice spacing through the use of small amounts of iodide, are in progress, and results will be reported soon.

Method of application. Although spraying and brushing were considered, a simple dip and drain with the slide held vertical for drainage has proved simplest. In the 30 seconds allowed for drying, the liquid collodion sets sufficiently so that it no longer leaves a wet mark when the edge of the slide is touched to absorbent paper.

If drying time is too short, the collodion will shrivel on contact with the silver-nitrate bath, and some of the collodion will wash off the slide and float on top of the bath. If drying is prolonged, the film will begin to dry hard, starting at the top. The metal halide will crystallize out and the film produced will be granular, noncontinuous, and wholly unsatisfactory.

Enough solvent must be left so that the metal halide is still in solution around the test specimen when the slide is put in the silver nitrate. The onset of drying at the top is not objectionable, but it may be taken as a warning sign to immerse the slide at once.

Silver-nitrate-bath composition and function. The purpose of the silver-nitrate bath is to provide a water solution of silver ions that can be taken up by the bromide ions in the collodion film to form silver bromide. Silver bromide, being highly insoluble in water, remains in the film. As the number of silver and bromide molecules in the film increases, crystals of silver bromide begin to form.

The action of the sulfuric acid in the silver-nitrate bath is somewhat subtle. Reducing the *p*H to 2.5 by the addition of sulfuric acid produces a film which is more sensitive and in which there is substantially less fog than in a film prepared without the acid. There is, in fact, a noticeable difference between *p*H 3 and *p*H 2.5. These differences are particularly noticeable when the exposure must be prolonged over many hours. On the other hand, a *p*H of 2 often results in an increase in fog over *p*H 2.5 after about 32 hours.

Originally, the practice in wet-collodion photography was to use nitric acid as a fog-suppressing agent. This was found to work only moderately well. Sulfuric acid was substituted in our studies in an attempt to lower the *p*H and to introduce an excess of sulfate ion to inhibit the action of the ferrous sulfate developer. The fog suppressing action of sulfuric acid proved to be far greater than that of nitric acid for the same solution *p*H. In addition, increased radiation response, possibly due to the silver sulfate formed, was noted. Thus, an increase in sensitivity and a decrease in fog have been achieved simultaneously.

Use of refrigeration. The most important factor in making the wet process of autoradiography useful has been temperature control for fog suppression.

Figure 2 shows three slides on which radioactive scandium oxalate was deposited and which were then autoradiographed. In processing slide 647, all solutions were at room temperature, about 24° C. The fogging, while extreme, is not a singular occurrence. Such fogging has been observed often in the wet process where no temperature control was used. Slide 641 was processed in a refrigerated silver-nitrate solution such as that outlined earlier. Almost complete fog suppression was achieved. There was no other difference in the processing of these slides.

If the developer is also refrigerated, it is not capable of bringing down an image in the time allotted for development. Since development is a chemical reaction depending on the random

velocity of ions in the solution, a reduction in temperature strongly inhibits the reaction. The effect of this is shown in slide 645.

The physical effect of the lower temperature, i.e., why it suppresses fog without inhibiting radiation response, has been the subject of intense study.

Actually, it is impossible to distinguish a "fog" grain from one reduced by radiation. A fog grain is simply one in which, by some undesirable combination of circumstances, the silver development centers have become large enough to precipitate more silver during development. Since the growth of the development centers is normally brought about by the action of radiation, there is no way to distinguish the effects of fog and those of radiation in the final film.

Fog control is partially accomplished by lowering the *p*H of the silver-nitrate bath and the developer. Lowering the *p*H of the developer by adding sulfuric acid changes the development or redox potential and makes the developer less active. The potential change is responsible for the reduction in fog achieved by this method.

Assuming the development centers built up by spontaneous or random action will be smaller than those produced by radiation, this technique of reducing development potential will help suppress fog. Unfortunately, the assumption that the fog specks will be smaller than the image specks is not always true; then no amount of acid will help. By using low temperatures (1° C) for the silver-nitrate bath, the basic cause of fog, the spontaneous growth of random centers, is inhibited by the reduction in ion mobility.

There are two possible mechanisms for the creation of centers. First, if the grain growth is rapid, there is a great possibility of introducing flaws and faults that can serve as development centers. By lowering the bath temperature from 24° C to 1.0° C, the mobility of the ions in the solutions and the rate of crystal growth are smaller. This has been observed in our electron-microscope studies of the change in the rate of growth with temperature. The slower growth results in a more perfect crystal, and hence fewer faulty or fog grains.

Second, there is the matter of the generation of fog by methods quite analogous to the formation of an image. The Gurney-Mott mechanism, an accepted theory of image formation, depends on two phases in image formation: (a) an electron released into the conduction band within the crystal by light is trapped at a sensitivity speck; (b) an interstitial silver ion migrates to the speck, captures the electron, and becomes a silver atom. This mechanism is discussed later.

Because of thermal agitation, there is always a supply of conduction electrons in a silver-bromide crystal at room temperature. These electrons can also become trapped by a sensitivity speck, neutralize an interstitial silver ion which drifts up, and so form a silver atom. If the time delay between trapping and neutralizing an ion is long enough, however, the electron will leave the trap and no silver atom will be formed. The preservation of film at low temperatures is due to the increased average time delay between trapping the electron and neutralization of a silver ion.

In ordinary film, fog is also suppressed by silver atom dissociation. In a silver-bromide crystal, a silver atom also has a finite life. The atom eventually loses an electron and is tied into the lattice as an ion. Unless the rate of silver-atom formation exceeds the dissociation rate, any image or fog fades. Lowering the temperature slows down the spontaneous ionization of the silver atoms, so that the image in exposed film, particularly nuclear-particle tracks,

can be preserved over long periods of time.

As indicated, it has been observed that lowering the silver-nitrate-bath temperature from 20° C to 1° C almost completely eliminates fog without noticeably affecting sensitivity.

It is believed that the elimination of fog has been brought about by the reduction of ion mobility at the lower temperature. Actually, ion mobility within the crystal drops by a factor of 5 for a temperature change of from 20° C to 1° C.

Development. As outlined earlier, the slide, having been brought to room temperature in the warm silver-nitrate bath, is transferred to the developer.

Physical development is used in the wet autoradiographic process as in the original wet-collodion process. The physical developer consists of a mixture of silver nitrate and a reducing agent, but the mixture is chemically adjusted so that the reduction of silver nitrate to metallic silver takes place slowly. However, the presence of metallic silver catalyzes the reaction and makes it take place rapidly. Thus, wherever a sufficiently large sensitivity speck has been formed, it is rapidly enlarged by the further deposition of silver from the developer. The size of the silver grains in the final image is determined not by the size of the silver-bromide grains deposited originally in the film but by the amount of silver deposited from the developing solution. This can be used to control the final image.

The reducing agent used in the physical developer is ferrous sulfate. The source of silver ions is the silver-nitrate solution that adheres to the slide when it is removed from the silver-nitrate bath. These two solutions mix when the slide is placed in the ferrous-sulfate solution, and then the actual physical development takes place. In some cases, silver nitrate is added directly to the developer solution; when it is, it is done immediately before the developer is used.

The film grains contain sensitivity specks, and these are enlarged by development until the image is visible. It is believed that about 200 silver atoms form a development center (*6*). An important question of control arises immediately. At what point does the enlargement of the image or sensitivity speck stop? What determines the silver-grain size of the final image?

The endpoint is determined by the change in redox potential which occurs as development proceeds. Development continues as long as a potential difference exists, in accordance with the following equation:

$$\Delta E = E_{Ag} - E_{Fe} \neq 0$$

The equation governing the development reaction is:

$$Fe^{++} + Ag^{+}NO_3^{-} \rightleftharpoons Fe^{+++} + Ag + NO_3^{-}$$

The equilibrium point of this equation lies far to the left. In other words, the reduction of only a small number of silver-nitrate molecules will bring the reaction to a stop. However, if it were possible to reduce the effective concentration of ferric ions, the redox potential, E_{Fe}, would be lowered, and the reaction would continue farther.

This can be done by introducing an organic acid, such as acetic acid, into the developer. The acetate forms a complex with the ferric ion but has very little effect on the ferrous ion. By complexing the ferric ion and so inhibiting the rise in the redox potential, the reduction of silver nitrate can proceed farther before equilibrium is reached.

Since the silver grain in the final image is formed by depositing silver on the sensitivity speck in the silver-bromide-grain surface, the size of the final silver grain is determined by the point at which equilibrium is reached. The grain size of the final image is determined not by the characteristics of the silver-bromide film formed during

the hours of exposure but by the developer used during the last few minutes of processing.

Where a set of tissue slides has been made from serial cuts on the same tissue block, it is advantageous to vary the grain size in successive slides. If activity is low, so that only a small number of silver grains is formed, it is useful to make these grains large, so that they are easily found under the microscope. Where activity is high, so that a large number of grains will be formed, it is best to keep these grains small, so that they do not obscure the tissue structure underneath. By changing the acetic acid concentration, the average grain size in the final films has been varied from 0.2 to 10 microns.

The organic acid concentration is not the only factor that controls grain size. The others are temperature, development time, and chemical concentration in the solutions. The acid concentration, however, is the most important single parameter and the one most conveniently varied.

Since the redox potential is critical in the behavior of the developer, changes caused by oxidation during storage are undesirable. To avoid such difficulties and obtain uniform results, it has been found best to mix small batches of developer just before use.

Fixing. After development, the slide is immersed in a solution of photographic hypo or sodium hypothiosulfate. In this solution, the unreduced silver bromide dissolves out through the formation of a silver ion-thiosulfate ion complex.

Metallic silver or silver atoms dissolve far more slowly, and so the silver bromide may be removed without affecting the silver image. Prolonged immersion in hypo, however, will weaken and finally dissolve the image. Since about 30 seconds is twice the time needed to remove the visible silver bromide, there is no need for prolonged immersion, nor is there much danger of forgetting a slide in the hypo.

Almost any commercial fixing solution can be used.

Quantitative Sensitivity and Grain-Size Distribution

The number of beta particles which must impinge on or pass through a grain to render it developable is one of the most important considerations connected with the sensitivity of an autoradiographic film. Investigations of single-grain films by Kinoshita in 1910 showed the number to lie between 5 and 10 particles from radium C (*7*). Yagoda estimates that 7 hits are required (*8*). Only in the past few years have films been produced that are sufficiently sensitive to record the passage of one electron.

In any emulsion, grains vary somewhat in sensitivity. Even where electron tracks can be detected, they are not continuous but are formed by an intermittent series of grains.

Judged from a picture of a 100-kv electron track published by Spence (*9*), the efficiency of grain production in NTB film is between 33–50%. About every second or third grain was rendered developable by the 100-kv electron. Since ionization by electrons of higher energy falls off until it reaches a minimum at around 800 kv, beta particles from such isotopes as iodine-131 and phosphorus-32 would be even less effective in producing grains than the 100-kv electron tested by Spence. Iodine-131 has a peak beta energy of 0.69 Mev and phosphorus-32 a peak energy of 1.6 Mev. Maximum beta-particle abundance occurs at about 0.3 Mev and 0.6 Mev, respectively.

The quantitative sensitivity of wet-collodion emulsions to beta particles from iodine-131 was determined using a modification of the method of Svedberg and Anderson (*10*). In 1921, they tested the sensitivity to alpha radiation

of single-grain layers made of haphazardly spread grains of uniform size. They found that the number of grains rendered developable followed Poisson's law of small numbers. In 1948, Webb and Evans checked these results and found no disagreement (*11*).

The Poisson law, as adapted to the problem by Webb and Evans, states that if a single grain of area a receives $a \cdot n$ quanta, on the average, the probability that it will receive j quanta is

$$P(j) = \frac{e^{-a \cdot n}(a \cdot n)^j}{j!}$$

where $P(j)$ is the probability of j hits on a grain of area a, n is the average number of quanta arriving per unit area, and e is the base of natural logs.

From this it may be shown that k, the number of grains per unit area receiving r or more hits is

$$k = N\left(1 - e^{-a \cdot n} \sum_{j=0}^{r-1} \frac{(a \cdot n)^j}{j!}\right)$$

where N is the total number of grains per unit area.

If one hit is sufficient to render a grain developable, this reduces to

$$\frac{k}{N} = 1 - e^{-a \cdot n}$$

This is the commonly used form and is applicable to alpha-particle studies where one hit is sufficient to render a grain developable.

To get the necessary data to evaluate beta-particle-induced response, uniform area sources of radioiodine with graded activities were prepared in a series of six slides. The slides were processed for two hours according to the directions given in the formula section. Three small disks of uniform area were cut from each autoradiographed source and were mounted on separate slides for study. Eighteen sources were made in this way, but three had to be discarded.

The area of the disks was known. By using an absolute beta-count technique, the source activity was found for each disk. The sources were then placed under the microscope at 400 diameters, and grain counts were made of randomly selected areas of known size. The silver grains were also classified by optical examination into sizes so that distribution studies could be made. These grains were the silver grains formed in development—not the original silver-bromide grains.

Over 17,000 grains were individually classified in this procedure. In any one disk, about $\frac{1}{250}$ of the total area was examined and counted. This was broken into five subareas across the disk diameter.

From this work, the number of grains rendered developable by a source with a specific disintegration rate was determined.

The Poisson small-number formula was used to evaluate the data and to find the value of r which gave the best correlation between the theoretical formula and the experimental results.

The heavy curves in Fig. 3 represent the Poisson formula for $r = 1$, 2, and 3, respectively. In addition, the experimental points are plotted, with a dotted curve connecting them. The data were corrected for background fog, so that the points represent only the number of grains rendered developable.

The apparent minimum number of hits required as determined from the

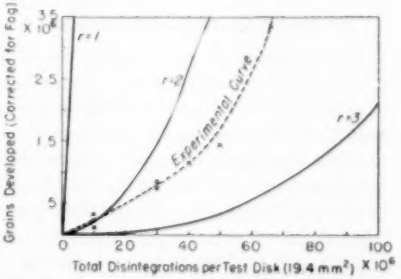

FIG. 3. Number of grains rendered developable vs total source beta emission as determined experimentally and by probability calculations

graph lies between two and three. However, a hit is not a well-defined quantity, since various amounts of ionization in a crystal can be produced, depending on the path of the electron through the crystal. It is, therefore, not surprising that the experimental curve does not coincide with a theoretical curve corresponding to a whole number of hits. The efficiency of the process is fairly satisfactory, although it would be more desirable for one hit to render any grain developable.

Actually, in the test films, short tracks indicating the passage of a single electron were observed. These tracks could be due to a particularly sensitive group of crystals, an electron of the proper velocity to produce maximum ionization, or the tracks could be artifacts. However, they have been observed too often to accept this last explanation.

These tests were made using iodine-131 as a beta-particle source. As shown by Steinberg and Solomon (*12*), the effectiveness of beta particles in producing photographic response varies with energy. The Poisson theory implies the use of particles of uniform energy, and so the results obtained represent an effect corresponding to an averaged energy. Individual electrons of critical energy could have efficiencies exceeding those of the averaged group, accounting for tracks found in the films.

Protective Coatings and Substrates

The possibility of direct chemical interaction between the solutions used in the wet autoradiographic process and the specimen being processed has already been considered. Where control specimens are not sufficiently reliable for indicating where silver has been deposited without the presence of radiation, more rigorous control procedures must be employed.

The most reliable autoradiographs are prepared under conditions where the test object and the autoradiographic layer cannot interact. This may be accomplished by interposing a thin chemically inert protective coating between the test object and the solutions. Ideally, such a coating would have zero thickness, be completely transparent to all radiation, and yet keep the solutions and test-object molecules far enough apart to prevent chemical interaction for an indefinite time interval.

No coating has been found which has the ideal characteristics listed above. Under the conditions encountered in the wet autoradiographic process, the best coating for use on tissue specimens found thus far falls far short of the specifications. A 1.5-micron (15,000 A.U.) layer has been found which will protect the tissue for about 4 hours. A 2.5 to 3-μ layer will protect it for 24 hours when application conditions are ideal.

It is obvious that the use of these protective layers must impair the resolution achievable in this system of autoradiography. Work is being continued to find thinner effective protective layers and thus maintain one-micron resolution in all cases. Vinyldene chloride in long-chain molecules, known commercially as "Saran 2000," has provided the best protective layers to date. The material is dissolved in methyl ethyl ketone and applied, by dipping, to special plastic slides on which tissue specimens are mounted.

A 2% solution (2 gm of Saran 2000 powder in 100 cm^3 of methyl ethyl ketone) leaves about 1 to 1.5 μ of protective plastic over a mounted tissue section. A 4% solution leaves about 2.5 to 3 μ. The collodion layer is applied over this protective coating.

Unfortunately, all the protective coatings of any value tested peel off a glass microscope slide when the coating is thin and the coated slide is immersed in water or weak acid (*p*H 2.5).

When the protective layer peels, it invariably takes the tissue specimen off the glass slide, because the tissue has become impregnated and imbedded in the plastic layer. By using a suitable plastic slide, intimate bonding between the plastic slide and the plastic coating can be achieved, and peeling becomes impossible.

Because of convenience and availability, celluloid slides have been used. However, other materials such as sheet polystyrene and polyethylene can be expected to do equally well or better.

For work with tissue specimens, Saran 2000 yielded the best results of ten materials tested.

For work with metal specimens mounted in Bakelite blocks, Vinylite VYNS gave a slightly superior protection than other materials.

Where solid metals are to be protected, there is no question of permeating the material. A solid homogeneous layer must be formed on the surface. Actually, both Saran and Vinylite have rather poor adhesion to metals, but excellent adhesion to Bakelite. The protection to the metal is essentially due to a very thin sheet of plastic anchored to the Bakelite mount around the edges of the metal specimen.

In applying Saran or Vinylite to a tissue slide, the slide is allowed to soak in the solution for one minute. The plastic actually penetrates and permeates the tissue so that after the plastic is dry, the tissue is literally imbedded.

The one-minute immersion is long enough for the solvent, methyl ethyl ketone, to attack the celluloid slide. However, on drying, a very hard, slick, and clean surface of Saran or Vinylite is formed on the celluloid. A clean surface is essential to prevent fog in the finished autoradiograph, and the new surface formed by the Saran or Vinylite is close to perfect. In addition, if the protection fails in a local area, the silver nitrate does not spread through the tissue, since the tissue is imbedded, not just covered. Failure at one point on a metal surface is usually more serious, since the adhesion between the coating and the metal is not very good.

The use of thermosetting materials, in connection with the possibility of soaking a tissue slide in a monomeric solution that could later be polymerized, was also studied. The small size of the monomer molecule permits complete penetration of the tissue. It was hoped that the subsequent irreversible polymerization of these monomers into a nonsoluble plastic would completely imbed and protect the tissue. Polymerization, once the proper chemicals are present, can be made to take place without heat or pressure simply by lowering the *p*H through the addition of acid to the solution. Before polymerization, the monomeric constituents are usually in water solutions.

Urea formaldehyde and resorcinal formaldehyde were tested for this purpose. The first tests yielded poor results, far inferior to those of the thermoplastics in organic solvents. However, it is believed that the principle involved is sound, and additional, more extensive tests are planned.

Sheet celluloid has been used for mounting tissue specimens, but testing to find a better tissue mount than celluloid is being continued. The celluloid cannot be used with alcohol-base tissue stains, since celluloid is soluble in alcohol. In addition, the celluloid slide tends to warp after prolonged immersion in the silver-nitrate solution. At present, tissue slides are stained using water solutions and the slides then stored in toluene. Toluene does not attack celluloid, but appears, on long immersion, to toughen it and make it somewhat less susceptible to attack by alcohol and methyl ethyl ketone.

The discovery of a thin protective coat with the necessary properties, and which could adhere to a glass slide,

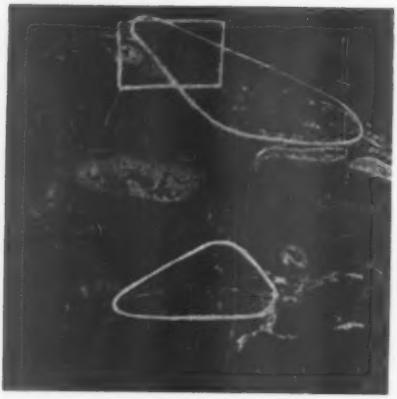

FIG. 4. Neck-tissue section of rat given I^{131}. Thyroid gland areas are outlined in white. Slide coated with radiation-sensitive layer. Direct projection. Fig. 5 shows upper outlined area enlarged

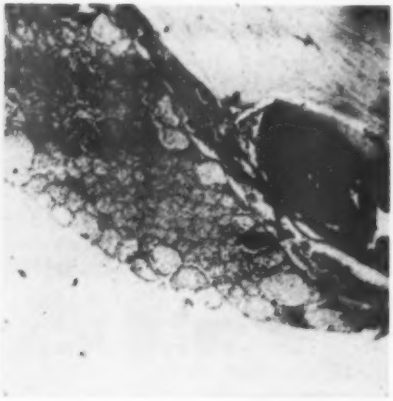

FIG. 5. Processing of this autoradiograph of the thyroid area outlined in Fig. 4 was adjusted to produce moderate silver grain size. Tissue was protected with one coat of 4% Saran 2000. I^{131} is apparently more concentrated in smaller acini toward center of gland lobe. Other larger acini are still filling

would eliminate the disadvantages of plastic slides and the restrictions on the type of stain used. However, until such a glass-adhesive coat is found, a superior plastic slide will be sought.

Examples of Wet-Process Autoradiographs

Wet-process autoradiography has been used, up to this time, only to study the distribution of radioiodine (I^{131}) in the thyroid gland of a rat, and the distribution of induced radioactivity in a high-cobalt metal alloy which had been exposed to the deuteron beam of the University of Michigan cyclotron.

The tests actually were intended to check the new method of autoradiography under fairly well-known conditions rather than to uncover new facts. Extensive use of the technique for study of new problems is planned. For purposes of this report, only the results with thyroid tissue will be reported.

The thyroid studies were made on the gland of a rat which had been given about 400 microcuries of I^{131} 24 hours before being sacrificed. Tissue slices 5 μ thick from the neck of the rat were prepared, mounted on celluloid slides, and then stained in an aqueous solution of haemotoxylin, a blue stain with a high affinity for cell nuclei. The prepared slides were stored in toluene until used.

To prepare an autoradiograph, a slide was removed from the toluene, allowed to dry slightly, and then immersed for one minute in a solution of Saran in methyl ethyl ketone. The slide was then dried to hardness under an infrared lamp. Drying was accelerated as much as possible in damp weather; otherwise absorbed moisture caused cloudiness in the plastic coat.

As a result of the plastic impregnation, the tissue was now completely

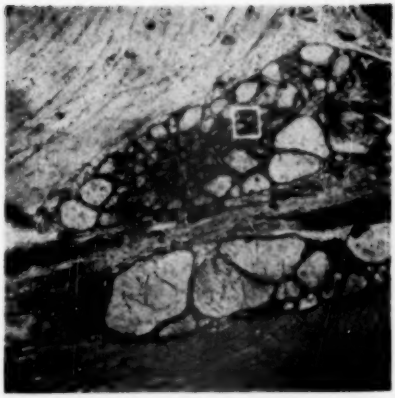

FIG. 6. Processing of this autoradiograph of thyroid tissue section was adjusted to produce moderate silver grain size. Tissue was protected with one coat of 4% Saran 2000. Concentration of I^{131} in smaller acini is very marked

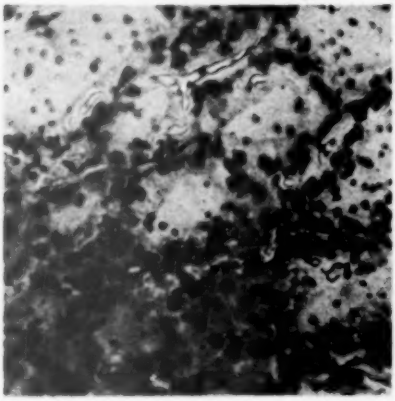

FIG. 7. Detail photomicrograph of area outlined in Fig. 6. Black spots are silver grains. Dark gray areas slightly larger than silver grains are the nuclei of acinar epithelial cells. Note clustering of grains in these cells

imbedded in an inert material and was histologically well preserved. Assays of the radioactivity in a tissue section were always made after this imbedding process, since the slide could be handled freely and there were no problems of the tissue drying out, absorption of beta radiation by excess toluene, or the effect of the toluene vapor on the Geiger-tube window.

Figure 4 is reproduced from a photographic enlargement of neck-tissue section of a rat. A tissue slide was used in place of a negative in the enlarger. This tissue section was completely processed for autoradiography. The silver deposit, due to radioactivity in the thyroid gland area, is too fine to be seen without the aid of color reproduction. The thyroid gland areas have been outlined in white.

Figure 5 is reproduced from a study at 100 diameters magnification of the upper outlined area in Fig. 4. The striate tissue adjoining the gland tissue is muscle and is seen to be practically free of black silver grains. The silver deposit in the gland area is quite heavy and correlates well with structure.

Thyroid-gland structure consists of small globules of a colloid material surrounded by a layer of cells, the epithelium. The gland is thus a series of closed thin-walled compartments or acini, each filled with colloid. Access to the colloid, in which the iodine in organic combination with protein (thyroglobulin) is stored, is obtained only by penetrating the epithelial layer of cells.

A thin section of the thyroid gland thus reveals a series of islands of colloid, each surrounded by a thin layer of epithelial cells. The smaller colloid pockets have a larger surface area per cm^3 of volume and, in these sections, seem to be located toward the center

of the gland lobe. The relatively dense deposit of silver in the central area of the gland studied indicates that most of the radioiodine was being taken up there, confirming the idea that iodine enters the colloid via the surface cells.

Some of the larger colloid pockets around the edge of the gland are particularly interesting in that they show a heavy silver deposit on the epithelial cells, but a much lighter deposit over the colloid. In these larger follicles, more distant from the center of the gland, the iodine is just making its way in via the epithelial layer. Close examination shows many short beta tracks.

The concentration of iodine in the center of a thyroid-gland lobe where the colloid pockets are small is shown particularly well in Fig. 6, which was reproduced from a 100-diameter autoradiograph. The larger acini are almost completely devoid of silver grains, indicating low radioactivity and hence low I^{131} content, while the small acini in the center of the gland lobe show a substantial uptake of the I^{131}.

Figure 7, reproduced from a 1,500-diameter photomicrograph, shows the relation between the silver grains in the wet process and the individual cells of the epithelium. It is the detail area marked in Fig. 6. In the technique used, the tissue section was not completely plane, and, consequently, there is some variation in definition as different areas lie in, above, or below the focal plane. Approximately in the center of the picture is a cluster of epithelial cells apparently well loaded with I^{131}. Some of it is in the process of passing into the colloid just above and to the left of the cell cluster.

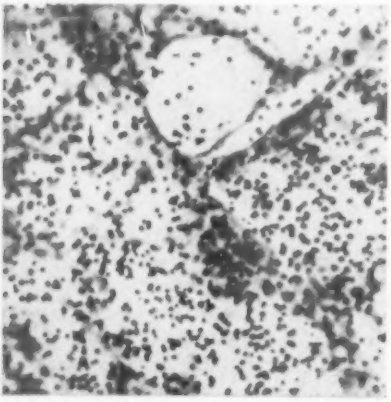

FIG. 8. Detail photomicrograph of thyroid-tissue section of rat treated with I^{131}. Arrow indicates two tracks believed to have originated in the same cell nucleus

Reproduced in Fig. 8 is a 1,500-diameter detail photomicrograph of an area of rat thyroid tissue section adjoining that shown in Fig. 5. Even though the focal plane in this photomicrograph is in the emulsion layer, tissue detail is discernible. There is an apparent concentration of I^{131} in the cluster of epithelial cells below and to the right of the center of the picture. The colloid area higher in the picture has not yet filled with a significant amount of I^{131}. Above and to the left of the epithelial cluster is an acinus in which the mechanism of transport of iodine to colloid through epithelium is demonstrated.

In the next pictures, grain size was varied by acetic-acid control. Where the acetic acid was omitted in the developer, the grains are much smaller than the cells, improving the study of fine detail substantially. In the lower left corner of Fig. 9 (reproduced from a 1,500-diameter photomicrograph), a curved line of silver grains is distinct in the inner margin of the epithelial lining. Presumably, this is due to elemental I^{131} in the cytoplasm of the epithelial cells as the process of storage in the colloid protein is progressing. This iodine was on its way into the colloid proper.

The very small grains are suitable for use in regions of moderate-to-high activity in order that their presence may be unambiguously established. In locations of lower activity where

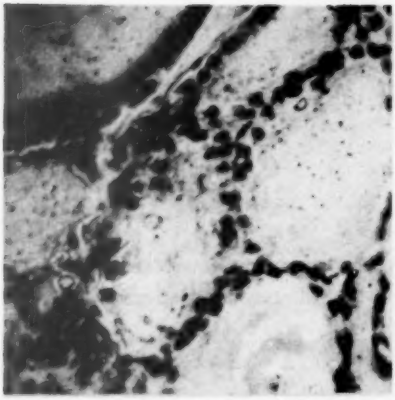

FIG. 9. Detail photomicrograph of autoradiograph of thyroid section processed for maximum detail. In lower left, note ring of grains due to I^{131} in cytoplasm of epithelial cells

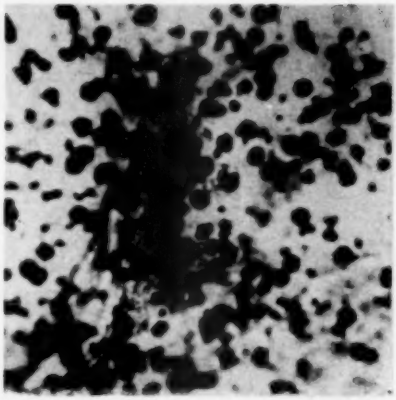

FIG. 10. Photomicrograph of autoradiograph of highly active site in acinus. Processing was adjusted to produce large grains of silver. Tissue covered with two 4% Saran 2000 layers, each ~2.5 μ thick

single grains may be significant, it is desirable to have much larger, easily detectable, grains. This has been carried to an extreme, again by acetic-acid control. See Fig. 10, which is from a 1,500-diameter photomicrograph. The large silver grains in the center completely cover a group of epithelial cells containing I^{131}. The picture presents no new histological information, only a change in technique.

Final Remarks

The wet process of autoradiography, while extending to about one micron the resolution possible with beta emitters, still leaves vast room for improvement. The identification of the atom from which a beta particle originated is the inherent limit of the radioactive tracer system. This awaits the creation of a detector system with one-angstrom-unit resolution.

* * *

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Deuteron Activation and Autoradiography as Coupled Techniques in Tracer Experiments

Variations from the usual steps of isotope addition and detection were employed in a tracer experiment revealing the behavior of sodium in refractory materials. Concentration and location of sodium in cross-section surfaces was shown by autoradiography of cyclotron-activated specimens

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THE TRACER TECHNIQUE used by the majority of investigators involves the addition of some foreign isotope to the reactants. This isotope nowadays is almost invariably secured from the Isotopes Division of the Atomic Energy Commission. After the reaction, the products are most generally measured by a counter in the case of radioactive tracers, or with a mass spectrograph for stable isotopes.

We have successfully departed from this usual procedure in the experiments to be described. The tracer isotope was made using the cyclotron of the Scaife Laboratory of the University of Pittsburgh. The accelerator was used to generate the tracer isotope from a component element of the sample after the experimental reaction to be followed was complete.

The problem under attack by this method involved the interaction of two materials, liquid glass and the various refractory materials that confine it in melting tanks and furnaces. The glass under consideration was composed mainly of silica, soda, and lime. The refractory materials were also mixtures of oxides, but had been selected to emphasize the property of high melting point. It is reasonable to expect that one set of oxides, the melted glass, would attack and dissolve the similar oxides of the container. This does happen and is of vital consideration both to the glass industry and to the manufacturers of refractory materials.

One important key to the mechanism of dissolution is the element sodium and its compounds. Sodium is used in glass primarily to facilitate the melting of the other materials. Unfortunately this property is still manifest after the glass is melted, so that the role of sodium in the reaction of hot glass on refractories is important. We were able to follow the migration of sodium in the refractories by the isotope tracer technique to be outlined.

The nuclear reaction was $Na^{23}(d,p)Na^{24}$, and took place in the external deuteron beam from the cyclotron (*1*). The deuterons, as they emerged through the thin aluminum window in the cyclotron vacuum wall, had energies of about 16 Mev. Since only a fraction of this energy was required to bring about the desired nuclear reaction, the samples were exposed at a suitable position a few inches from the end of the traverse of the deuteron beam in air. Here the

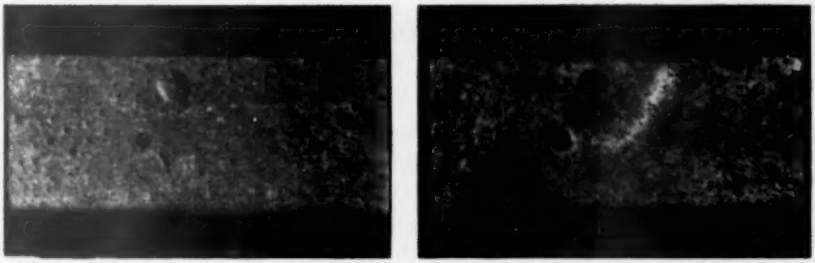

FIG. 1. Conventional (left) and radiation photographs of an unused refractory material. Autoradiograph shows location of sodium activated in cyclotron

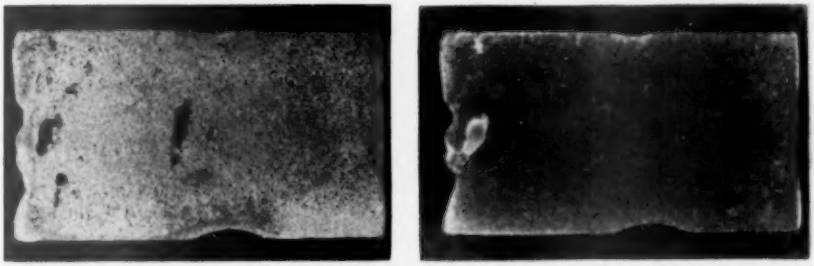

FIG. 2. Conventional photograph (left) and autoradiograph of another specimen of refractory material. Concentration of sodium around edges is due to infiltration from a molten soda glass at 2,650° F in which the specimen had been immersed

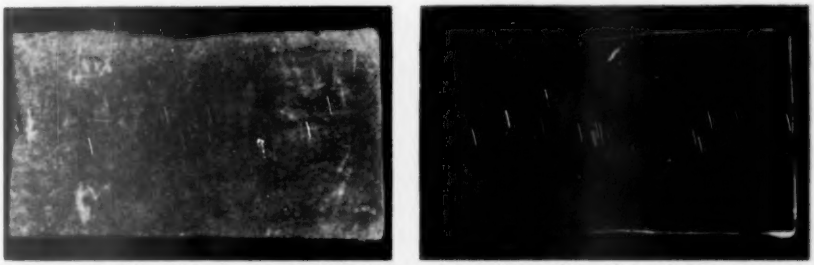

FIG. 3. Similar pictures, conventional then autoradiographic, of a refractory sample that did not pick up sodium from a glass bath. The white edge is the sodium in a film of glass, which has also filled a crack in the refractory

beam had a relatively large area and was quite homogeneous with a deuteron energy of 4–5 Mev. Sample position for irradiation can be seen in the picture on the front cover.

Irradiation times varied to some extent, governed by previous experience with the particular samples involved. In a typical run, the current collected by sample and sample holder from the beam might have been 10 μa, and the time of irradiation, one-half hour.

Sodium has a large cross section for deuterons, so the desired reaction was favored (*2*, *3*, *4*). Some side reactions did occur, probably resulting in the formation of 2.4-min Al^{28}, 112-min F^{18}, and 170-min Si^{31}. These isotopes were

not definitely identified, but account for the observed high initial activity. After 2 or 3 hours, however, the remaining activity decayed at the 15.1-hr rate of Na^{24} (*6*), as well as exhibiting the particle and quantum energies to be expected. Long-lived activities could hardly be significantly produced in such relatively short bombardments.

The result, then, of this irradiation was the radioactivation of a fraction of the sodium atoms in the surface layers of the sample material. The sodium content and distribution could then be determined by the detection of the emissions from the unstable tagging atoms. Since the collision of a bombarding deuteron with a sodium nucleus was a rare and random event, it depended uniquely on the number of atoms in the vicinity available as targets. Thus the atoms whose nuclei were hit, and thereby radioactivated, were distributed in the surface layers strictly in proportion to the local concentrations of all sodium atoms.

Three emissions result from the decay of Na^{24} to Mg^{24}. One is a beta particle of 1.4-Mev energy. Then, simultaneously, gamma rays of 2.8 and 1.4 Mev are radiated. Both types of emission influence the sensitive silver bromide grains in the photographic emulsion used to record those radiations, rendering the bromide grains developable.

Exposure of the plate or film was accomplished simply by placing the active face of the sample in contact with the emulsion layer. Duration of exposure depended on the original activity level of the sample. Most often, substantially all of the emissions were utilized by exposures of three or more half-lives.

Many investigators employing such autoradiographic techniques use X-ray or "nuclear" film because of its high sensitivity. However, with the cyclotron activation method described, it was comparatively easy to achieve relatively high activity levels. Hence, a slower plate of much higher resolution and small grain size was used (*5*). This was Eastman's SA-1, a plate designed for spectrophotography. The resulting images were suitable for analysis on a microphotometer of the type used for recording spectrum line intensities. Concentration as well as location of sodium could thus be determined.

For this particular investigation, such exact quantitative information was not necessary. It was sufficient to be able to judge the gradient of sodium concentration visually from the liquid-solid interface toward the interior.

Three typical autoradiographs, together with corresponding conventional light photographs, constitute Figs. 1, 2, and 3. The radiographs were reversed in printing in the usual photographic way; thus a white area in the figures corresponds to areas relatively high in sodium content. The range of concentrations displayed is from a fraction of a per cent to a few per cent of sodium. The concentration of radioisotope is, of course, many orders lower than this.

The samples illustrated were 2½ by 4½ in. sections of three different refractory materials. These had been made by melting mixtures of refractory oxides in an electric furnace, and then casting the molten mass into molds.

* * *

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Detection of Small Fluid Flows by Means of P^{32}

Preliminary experiments with a tracer technique show that, compared to the conventional visual method and a thermal method, it is the most sensitive method of detecting small fluid flows. The tracer method can also be applied to investigation of local settling in unconsolidated porous media

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EXPERIMENTAL researches on initial convective flow of fluids in porous media have customarily been based upon visual observations (*1, 2*). Having dissolved a small amount of dye in the fluid in such a way as to produce strong gradients in density of color, the experimenter observed the dye through a microscope and so inferred motion.

Although this method is capable of refinement by the use of continuously sensitive photometric apparatus instead of the eye, it is subject to one essential limitation: In order that the dye be visible, in a porous medium such as sand, it must be intimately adjacent to a window. Hence an observer could not by these means know precisely what was happening at points typical of the whole field of flow; at most, he would be limited to knowledge of occurrences at non-typical points adjacent to his window. While this limitation may not be serious in studies of large flow rates, it is inadmissibly severe in observations of small (as incipient convective) flows. On the other hand, the visual method has produced data in general agreement (*2*) with extant theory, and for simplicity probably cannot be surpassed.

If one were to use a radioactive substance as a flow indicator, detecting activity by means of a counter either in the fluid or outside of it, he could obtain data as to flow at points relatively remote (i.e., by many grain diameters) from the detecting element (*3*). Gemant has reported a technique (*4*) using this general principle for coefficients of diffusion in liquids, and the method has been used for studies (*5*) of diffusion in metals.

Let the fluid in its porous medium be locally inoculated with a small amount of purely beta-active substance, at an "effective" distance ρ from the thin wall of a submerged counter. Let $F(E)$ represent the energy-distribution function for the active material, in terms of kinetic energy E; and let $E = E(R)$ be "mean" energy-range relation for beta-particles in fluid and porous medium *in situ*. Then the counter's rate of counting will be

$$I \sim (1/\rho^2) \int_\rho^\infty F[E(x)][dE(x)/dx]\,dx \quad (1)$$

No account is taken here of straggling, scattering, finite volume of inoculant, inevitable bremsstrahlung radiation, or counter-wall thickness. In terms of fractional alterations of counting rate by an alteration, $\Delta\rho$, of distance ρ,

$$\frac{\Delta I}{I} \sim -\left\{2 + \frac{\rho F[E(\rho)][dE(x)/dx]_{x=\rho}}{\int_\rho^\infty F[E(x)][dE(x)/dx]\,dx}\right\} \times \frac{\Delta\rho}{\rho} \quad (2)$$

Clearly a small fluid flow should cause a $\Delta\rho/\rho$, and this would strongly affect

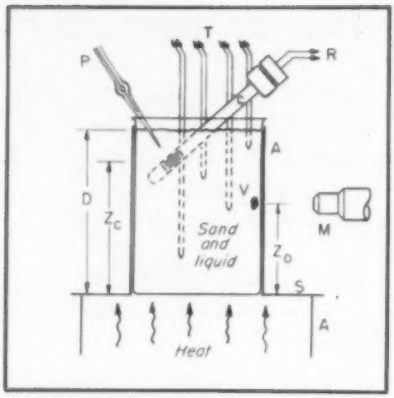

FIG. 1. Apparatus used for detection of incipient convective flow in porous media

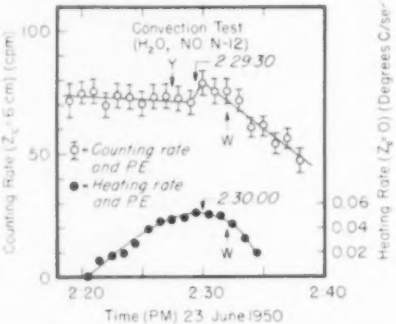

FIG. 2. Data obtained in test number N-12. Heat was applied at time 2:17:30. Only diffusive motion of dye was visible at time *Y*, while convective flow was clearly visible at time *W*

I, for $\Delta I/I$ is made up of two terms *of the same sign:* (1) a geometrical one $(-2\Delta\rho/\rho)$; and (2) another dependent upon ρ, $F(E)$, and $E = E(R)$ (the rest of the right member of Eq. 2). A high sensitivity was thus anticipated for the method before trial.

In using this principle, we have carried out convection experiments with the apparatus sketched in Fig. 1. The counting tube *C* was a mark 1, model 100 medical type manufactured by Radiation Counter Laboratories, Chicago, Ill. It was connected at *R* to a scaling and autographic rate-recording system. In Fig. 1, *A* is asbestos insulation, *S* is copper screen, *V* is methyl orange dye, *M* is a microscope used for visual observation of the dye, *P* is a micropipette in a typical location for inoculation, and *T* are thermocouples. Only four of the seven thermocouples used are shown.

After inoculation with several λ's of dilute $H_3P^{32}O_4$ solution ($\sim$0.01 to 0.1 μc), near, but not immediately adjacent to, the counter wall, the micropipette was withdrawn and counting was started. After withdrawal, settling occurred as the unconsolidated medium (sand) returned to stability, and, of course, diffusion occurred. These processes were watched in the counting rate, until it was judged that settling had ceased (nonfluctuating counting rate), at which time heat was applied from below until convection began. Convection was presumed to have started when a new trend of counting rate was established. It was found in general that such new trend was preceded by a sudden significant alteration in rate.

Figures 2 and 3 show typical data. Figure 2 refers to an experiment in which the total activity was small but settling had been allowed definitely to cease. At time 2:29:30 $\pm$ 00:00:30 a sudden increase in rate occurred, followed by a relatively rapid decreasing trend; this time was taken to be that at which convective flow began.

Figure 3, on the other hand, refers to an experiment with a larger inoculant activity, in which settling was continuing. At time 9:09:45 $\pm$ 00:00:30 a violent decrease in rate occurred, followed by a recovery and then a decreasing trend; this time was taken to be that at which convective flow began.

The two cases shown in Figs. 2 and 3 illustrate relative extremes among typical data. They suggest a most preferred type of experiment, in which:

1. There is no evidence of settling, as in Fig. 2.

2. The counting rate is high enough to have statistical significance, as in Fig. 3.

3. The start of flow is indicated by the beginning of a new trend in rate, preceded by a sudden alteration with or without recovery.

Just such a preferred experiment is illustrated by Fig. 4 (*6*); in it, flow was presumed to have started at **2:40:40** ± 00:00:25. Experience suggests that such excellent records are relatively difficult to obtain, and that Figs. 2 and 3 are obtained more commonly.

Twelve experiments of this type were carried out, each with somewhat different features; in each, determinations were made as to times of initial flow by visual and by thermal (*3*) means. Volume of inoculant was thus found to be significant, for volumes of 30λ or less usually produced the sharpest indications of flow. Sensitivity of the method does not depend upon the total volume of test fluid, however, if the fluid fully permeates the sand and if the volume of the counter is only a fraction of a per cent of that of fluid and sand.

Site of inoculation was found to be significant; too distant sites produced low counting rates, and too close sites led to insensitivity (probably because of peculiar flow patterns around the counter wall).

Location of counter was not significant, whether inside or outside the vessel of Fig. 1. But successful use of a counter external to the fluid vessel was found to be limited to tests in which inoculation was within about 1 cm of the vessel wall. Even then, the nearest portion of the counter wall should not be more than 3 or 4 cm from that point of the vessel wall nearest the inoculation. Vertical location of the inoculation, however, has no effect.

An auxiliary experiment* with Zn^{65} showed relative insensitivity, because the second term in the right member of Eq. 2 essentially vanishes for such a gamma-active material.

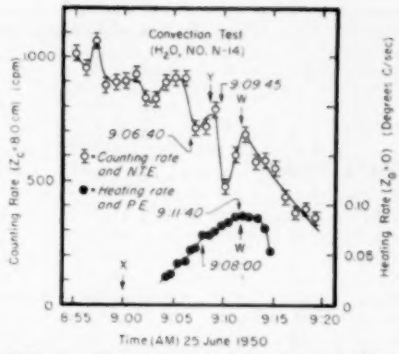

FIG. 3. Data obtained in test number N-14, in which severe settling occurred. Heat was applied at time *X*. Only diffusive motion of dye was visible at time *Y*, while convective flow was clearly visible at time *W*. The large settling, indicated in counting-rate change at 9:06:40, may have had a corresponding indication in heating-rate change at 9:08:00

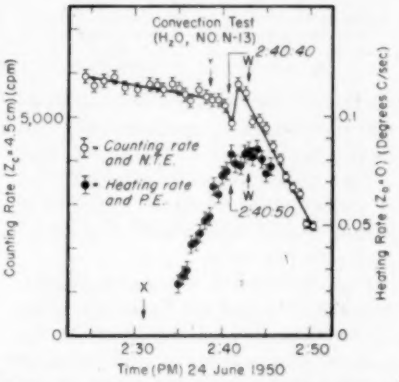

FIG. 4. Data obtained in test number N-13. Heat was applied at time *X*. Only diffusive motion of dye was visible at time *Y*, while convective flow was clearly visible at time *W*

The table on the next page lists for each experiment performed with P^{32} the times elapsed between application of heat and beginning of convective flow, the latter determined by each method

* The collaboration of Dr. H. D. Bruner, Oak Ridge Institute of Nuclear Studies, in this experiment is much appreciated.

Experiments with Water in Sand, Using P^{32} Indicator

Test	λ's inoculant	Permeability (c.g.s.)	Depth* (cm)	τ† (cm)	Time for detection of convective flow (min, sec; % visual) Visual Method		Thermal Method		P^{32} Method	
N-8‡	100	256×10^{-8}	11.7	0.8	11:15	100	8:00	71	7:40	68
N-9	5	433	13.1	1.0	8:30	100	6:00	71	6:00	71
N-10	10	433	12.2	1.4	8:30	100	9:00	106	§	§
N-11	15	433	12.2	1.9	7:30	100	9:30	127	§	§
N-12	10	433	11 ± 0.5	0.9_5	14:30	100	12:30	86	12:00	83
N-13	75	433	10.3	0.9_5	12:00	100	9:50	81	9:40	80
N-14	60	433	11.9	0.9_5	11:45	100	11:40	99	9:45	83
N-15	160	433	11.6	1.2	11:00	100	9:15	84	¶	¶
N-16	100	433	11.4	1.3_9	13:30	100	11:25	85	10:45	80
N-17	200	433	11.2	1.2	13:00	100	9:30	73	11:30	89
N-18	30	433	11.5	1.2	12:00	100	10:00	83	11:20	95
N-19	75	433×10^{-8}	10.6	1.1	12:15	100	12:00	98	11:10	91
Average						100		89		82

* Distance given is depth of water and sand *in situ*.
† Temperature decreases from value at bottom of vessel, by amount T_D at the top of sand and water. τ is distance, from bottom, through which temperature drops by amount $(1 - 1/e)T_D$.
‡ Counter outside of vessel.
§ Excessive settling, continuing during initial flow.
¶ Inoculation in contact with counter wall.

mentioned previously. The radioisotope method gave, generally but not always, the earliest indication of flow; i.e., the method was the most sensitive for detecting small flows. This is in general accord with qualitative expectations derived from Eq. 2.

It is appropriate to mention, moreover, that comparisons have been made (7) of thermal environments at the start of flow against those predicted by extant theory (8). Average deviations between theory and experiment were 150% for visual detection, 32% for thermal, and 11% for radioisotopic. Although the theory is not yet rigorously correct, for which reason the matter is not further discussed here, these average deviations point to the same conclusion as does the table.

An interesting by-product of Eq. 2 and the radioisotope technique as described, is their possible use in studies of local settling. Thus in experiment number N-14, illustrated in Fig. 3, the following maximum-to-minimum per cent changes can be attributed to settling: +12 at 8:57, −18 at 8:58, −12 at 9:02, +12 at 9:03, −25 at 9:07, and +10 at 9:08. The mean absolute value of these is 15%, which would correspond to 7.5% changes in ρ if the shape of the beta spectrum of P^{32} were ignored. The shape term in Eq. 2 can be estimated by mean values, as

$$\frac{\rho F[E(\rho)][dE(x)/dx]_{x=\rho}}{(\rho - \rho_{max})\bar{F}[E(\rho_{av})][dE(\rho_{av})/d\rho_{av}]}$$

where $0 \leq \rho_{av} \leq \rho$; in order of magnitude it is $(1/2)(3/1)(2/1) = 3$. Hence 15% fluctuations in $\Delta I/I$ can be attributed to 3% or 0.03 cm fluctuations in $\Delta\rho/\rho$, for $\rho \sim 1$ cm in all these experiments. Sands used in these experiments were composed of irregularly shaped grains with mean diameters of about 0.1 cm, so that the settling in experiment number N-14 was in amounts of somewhat less than one grain diameter. This approach to investigations of local settling is obvi-

ously susceptible to considerable refinement, as by the simultaneous use of three or more counters, and appears potentially very fruitful.

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The Linear Electron Accelerator as a Pulsed Neutron Source

With a pulse of 1 μsec and current peaks up to 100 ma, the linear electron accelerator can give large yields of pulsed photoneutrons. Preliminary estimates of source strength and investigations of the possibilities of its use are described

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TWO FEATURES of the linear electron accelerator render it uniquely useful as a neutron source for a time-of-flight velocity selector. The first feature is that it is a pulsed machine whose pulse has the relatively short duration of about 1 μsec. The second feature is that it is capable of delivering very high peak electron currents, up to about 100 ma.

For the bremsstrahlung gamma radiation obtained from electrons of about 20 Mev, the photoneutron yield is large. Thus the high electron currents are capable of yielding short neutron pulses whose intensity is greater than those available from pulsed cyclotron beams by several orders of magnitude.

The use of a linear electron accelerator as a pulsed neutron source was suggested by Cockcroft (*1*). The possibility of such use was demonstrated by Duckworth and Merrison (*2*) who produced neutrons by irradiating a heavy-water target with gamma rays produced by stopping electrons in a lead target. For a pulse of about 2 μsec, having a peak current of about 100 ma and an energy of 3.2 Mev, they obtained peak yields of about 2×10^{12} neutrons per second.

The use of this source in a time-of-flight neutron velocity selector has been described by Merrison and Wiblin (*3*). The gamma rays, produced by electron irradiation of a water-cooled Pt target,

TABLE 1
Resolution Factors for Some Time-of-Flight Velocity Selectors

Location	*Neutron source*	$\Delta t/L$ ($\mu sec/m$)
Columbia	Cyclotron	1
Argonne (fast chopper)	Pile	0.6
Harwell	Linear electron accelerator	0.5

produce neutrons in a mass of Be. The neutrons, after being slowed down by paraffin, traverse a 10-meter evacuated flight path and are detected in a 2.5-in.-diameter, 50-cm-long, proportional counter that has been filled with enriched BF_3 at a pressure of 60 cm of Hg.

The use of pulsed neutron sources for time-of-flight neutron velocity selection has been extensively described in the literature (*4*). The usefulness of a given spectrometer at a given neutron energy is measured by the resolution, which is the spread in the energies of the neutrons accepted by the detector at a given (energy) setting of the timing system.

For time-of-flight velocity selectors, the resolution is essentially determined by two factors. The first factor is the time spread of the pulse of neutrons from the source and of the interval during which the detector is sensitive. The second factor is the length of the flight path between source and detector.

The time spread, Δt, depends on the inherent time spread of the pulse of accelerated particles, on the "detector-on" interval, and on the spread in the times required to slow down the neutrons after production. The choice of path length, L, is presumably at the discretion of the designer of the velocity selector; however, the maximum L is clearly determined by the available neutron intensity (and the backgrounds) through the requirement that it shall be possible to gather the required data in a reasonable time.

The ratio $\Delta t/L$ is a convenient measure of the inherent resolution possibilities of a given velocity selector. Approximate values of this ratio for the three "best" available velocity selectors are given in Table 1. We note that $\Delta t/L \approx 1$ implies a resolution of $\Delta E \approx 1$ kev at $E = 1$ kev.

It is evident that improvements in resolution can be achieved both by minimizing Δt and by maximizing L. The most promising recent developments in both these directions have come from the attainment of pulsed electron sources of high energy and large peak currents (the Harwell velocity selector).

For electrons of <10 Mev, appreciable neutron yields can only be obtained through (γ,n) reactions on D and Be. However, for energies of about 20 Mev, or greater, it is possible to consider photoneutron reactions on other, heavier, elements. It is the purpose of this report to evaluate the possibility of utilizing the strong (γ,n) resonances observed in many elements at energies of about 15–20 Mev, to provide intense pulsed neutron sources.

Estimate of the Source Strength

For the purposes of calculation, tho following experimental situation is assumed. A beam of 20-Mev electrons falls on a thick block of some material, and the electron energy is converted into gamma radiation. The spectrum of the gamma radiation is given by $N(E_\gamma,E_0)$, which is the number of gamma rays per unit energy interval with energy E_γ for a single incident electron of initial energy E_0. The neutrons are produced by a (γ,n) reaction whose cross section, $\sigma(\gamma,n)$, is a function of the γ-ray energy. The γ-rays are absorbed through the usual processes (mainly pair production and

Compton scattering, at the energies of interest). The absorption cross section, σ_a, is also a function of E_γ.

If all the above defined quantities are known, the thick-target neutron yield can be computed as follows. Define a neutron-production cross section per incident electron as

$$\sigma(e,n) = \int_0^{E_0} \sigma(\gamma,n) N(E_\gamma,E_0)\, dE_\gamma \quad (1)$$

Since the γ-ray absorption is almost independent of E_γ (it does not vary much in the energy region of interest, 10 Mev $< E_\gamma <$ 20 Mev), an effective γ-ray absorption cross section, $\bar{\sigma}_a$, can be defined. The thick-target neutron yield, per incident electron, is

$$y = \sigma(e,n)/\bar{\sigma}_a \quad (2)$$

The rate of neutron production is

$$Q = y\Phi \quad (3)$$

where Q is the number of neutrons per second, and Φ is the number of electrons per second.

The next step is to estimate the cross sections.

Photoneutron cross section. In Fig. 1 the observed values of $\sigma(\gamma,n)$ are plotted against E_γ for Cu^{63}, Ag (Ag^{107} and Ag^{109}), and Ta^{181}. The first two are due to Diven and Almy (*5*), and that of Ta to McElhinney *et al.* (*6*). These were obtained by observing the activity resulting from the (γ,n) reaction induced by the X-ray beam from the Illinois betatron. The variation with energy was obtained from the observation of curves of the variation of induced activity with energy of the electrons in the betatron.

Three such excitation functions are shown in Fig. 2. The ordinates are in arbitrary units, different for the three curves. The interpretation of such curves, to yield the cross sections shown in Fig. 1, requires a knowledge of the X-ray spectrum from the betatron. Because the steps to go from Fig. 2 to Fig. 1 involve considerable uncertainties, the cross section values are probably uncertain by a factor of about 2. Furthermore, the results of different investigators may not be strictly comparable. Nevertheless, we shall, in the following computations, take these curves at their face value.

All of the curves of $\sigma(\gamma,n)$ show rather broad resonance shapes in the region of interest. There are two pos-

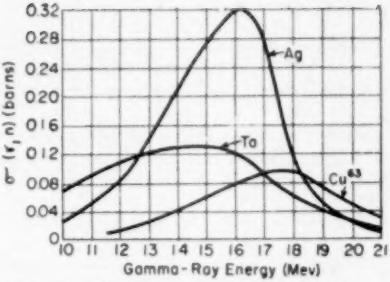

FIG. 1. Cross sections for the (γ,n) reactions on Cu^{63}, Ag, and Ta as functions of the gamma-ray energy

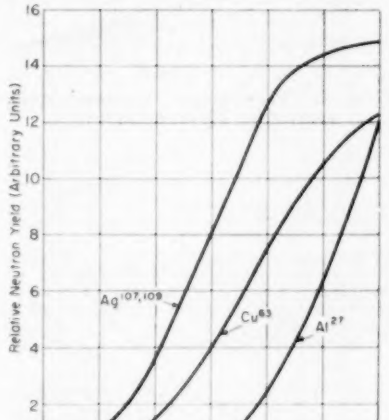

FIG. 2. Excitation functions for the (γ,n) reactions on Al, Cu, and Ag for thin-target bremsstrahlung gamma rays as a function of the energy of the electrons striking the target. The ordinates are in arbitrary units

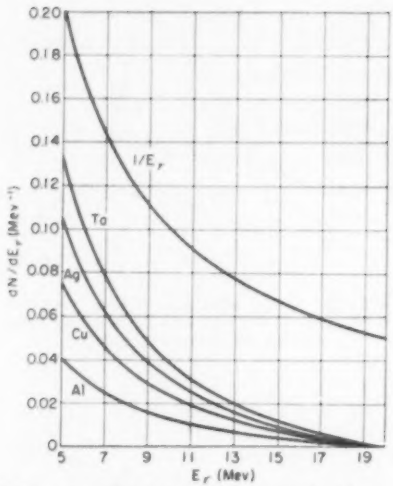

FIG. 3. Thick-target bremsstrahlung spectra for 20-Mev electrons

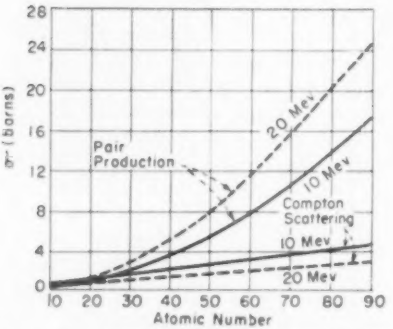

FIG. 4. Compton scattering and pair production cross sections per atom for gamma rays of 10 and 20 Mev

sible explanations for these resonances. (The actual reason might be a combination of the two.) The first explanation is that the peaks in $\sigma(\gamma,n)$ correspond to resonances in the cross section for capture of the γ-ray, possibly due to a dipole resonance process of the type suggested by Goldhaber and Teller (7). The second explanation is that these are not resonances at all but, rather, the falling off of $\sigma(\gamma,n)$ resulting from competition with a $(\gamma,2n)$ process at energies above the threshold for the latter.

If the observed shapes of the $\sigma(\gamma,n)$ curves arise from the second possibility, the neutron yields will actually be much larger than those computed on the basis of the curves of Fig. 1. However, the following estimates are based only on the (γ,n) reaction, and therefore will, if there is a significant $(\gamma,2n)$ contribution, represent underestimates of the neutron yields.

Thick-target bremsstrahlung spectrum. The γ-ray spectrum in the neutron-producing block of material is estimated according to the following approximate theory. The incident electron loses energy by radiation and by ionization. The rates of energy loss are assumed to be (neglecting a slowly varying logarithmic factor)

$$-(dE/dx)_r = aE$$

for the loss due to radiation, and

$$-(dE/dx)_i = b$$

for the loss due to ionization, where a and b are constants. Thus, the total rate of energy loss is

$$-dE/dx = aE + b \qquad (4)$$

After traversing a thickness x, the electron energy is given by

$$\ln[(aE_0 + b)/(aE + b)] = ax \qquad (5)$$

where E_0 is the initial electron energy.

The electron radiates as it loses energy. Assume that an electron of energy E, on traversing a thickness dx, gives off the bremsstrahlung spectrum of γ-rays

$$dN(E_\gamma,E) = (1/E_\gamma)(dE/dx)_r \times (1/E)\,dx = (a/E_\gamma)\,dx \qquad (6)$$

where E_γ ranges from 0 to E. Then, the total number of γ-rays per unit energy interval, at the energy E_γ, is

$$dN/dE_\gamma = N(E_\gamma,E_0) = \int_0^{x(E_\gamma)} dN(E_\gamma,E) = (a/E_\gamma)x(E_\gamma) = (1/E_\gamma)\ln[(aE_0 + b)/(aE + b)] = (1/E_\gamma)\ln[(E_0 + k)/(E_\gamma + k)] \qquad (7)$$

where

$$k = b/a = f(Z) \cong 814/Z \qquad (8)$$

TABLE 2
Thick-Target Neutron Yields for 20-Mev Electrons

Target element	Z	(γ,n) *threshold* (*Mev*)	$\int\sigma(\gamma,n)dE_\gamma$ (*Mev-barns*)	$\sigma(e,n)$ (*millibarns*)	*Effective* E_γ (*Mev*)	$\bar{\sigma}_a$ (*barns*)	*Yield* ($y \times 10^3$)
Cu^{63}	29	11.5	0.5	2.4	15	3.5	0.7
Ag	47	9.5	1.6	16	14	7.8	2.0
Ta	73	8.0	1.0	19	12	16	1.2

Equation 8 is obtained on the assumption that $b \propto Z$ and $a \propto Z^2$, neglecting the slight logarithmic energy dependence; the constants of proportionality are taken from Heitler (*8*). Equation 7 is probably reasonably accurate for $E_0 = 20$ Mev, in the energy range 10 Mev $< E_\gamma <$ 20 Mev. Figure 3 shows the thick-target γ-ray spectra for Al, Cu, Ag, and Ta.

Gamma-ray absorption. In the energy region of interest, the γ-ray absorption cross section is due to two processes, Compton scattering and pair production (*8*). The cross sections per atom for these processes are plotted separately in Fig. 4 as a function of the atomic number, Z, for γ-ray energies of 10 and 20 Mev. The sums of the absorption cross sections for the two processes are plotted in Fig. 5.

It is seen that the absorption cross section is not very strongly dependent on the γ-ray energy and that, for relatively heavy nuclei ($Z > 50$), it is primarily due to pair production.

Thick-target neutron yields. The data necessary for the computation of the number of neutrons per incident electron have now been assembled. Equation 2 is used for the computation, and the results for Cu^{63}, Ag, and Ta targets are given in Table 2. The values of $\sigma(e,n)$ are found from Eq. 1 with $E_0 = 20$, and are computed numerically from the curves of Figs. 1 and 3. The effective γ-ray energies are taken to be the energies at which $\sigma(\gamma,n)N(E_\gamma,20)$ have their maximum

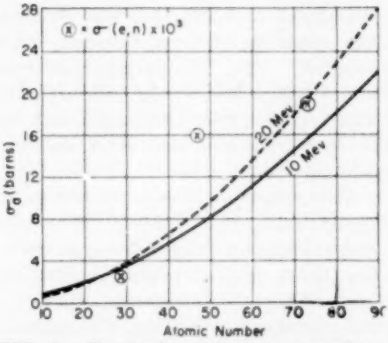

FIG. 5. Total absorption cross sections of 10- and 20-Mev gamma rays as functions of atomic number. The crosses are the computed (γ,n) cross sections, integrated over the thick-target bremsstrahlung spectra

values, and the corresponding values of $\bar{\sigma}_a$ are obtained by a linear extrapolation between the curves in Fig. 5.

Consider an electron current having a peak value of 100 ma and a duration of 1 μsec, repeated at the rate of 120 pulses/sec. Then

$$\Phi = 6.24 \times 10^{17} \times 10^{-6} \times 120 = 7.5 \times 10^{13} \text{ electrons/sec}$$

For a thick silver target, the expected neutron yield from this current is

$$Q = 2 \times 10^{-3} \times 7.5 \times 10^{13} = 1.5 \times 10^{11} \text{ neutrons/sec}$$

Preliminary Experiments

During the past year there has been in operation at MIT a linear electron accelerator. The essential features of its design have been described by Slater

(9), and the details of its construction and operation have been reported by Demos (10). As currently operated, the accelerator yields 16-Mev electrons in 2-μsec pulses; the peak current is about 10 ma with a repetition rate of 120 cps, and the average current is about 1.2 μa.

The neutron yields, for electrons impinging on thick targets of various materials, have been measured by Burkhardt and Fox (11) who detected the neutrons with a shielded "long counter" (12). The targets used were aluminum, brass, lead, and uranium. The neutrons from the target were collimated by a hole in a thick lead and paraffin shield.

The results of these measurements are summarized in Table 3. The theoretical value for brass was computed by the theory outlined in the previous section. For this calculation, the (γ,n) cross section for Cu^{63} and for electrons of $E_0 = 16$ Mev was used.

Further experiments on the neutron yields and energy distributions, and continued exploration of the velocity-selector possibilities of the accelerator, are being carried out by the accelerator group under the direction of I. Halpern.

TABLE 3
Thick-Target Neutron Yields for 16-Mev Electrons

	Yield (neutrons/electron)	
Target	*Observed*	*Calculated*
Aluminum	1×10^{-5}	
Brass	1×10^{-4}	1.3×10^{-4}
Lead	1×10^{-4}	
Uranium	1.5×10^{-4}	

Velocity-Selector Possibilities

A number of neutron characteristics pertinent to the choice of a neutron source for a velocity selector are given in Table 4. The fourth column of the table lists the average number of collisions with hydrogen that a 2-Mev neutron would have to undergo to reduce its energy to that indicated.

Table 4 also shows that a 100-kev neutron, for instance, takes 4.6 μsec to travel 20 meters. Thus, if the time resolution of the velocity selector can be kept to $\lesssim 1$ μsec, it should be possible to make measurements, with fair resolution, up to about 100 kev.

The main problem is one of intensity. For a source of about 2×10^{11} neutrons per second, the flux at 20 meters is

$$\phi_n = 2 \times 10^{11}/4\pi \times 4 \times 10^6 = 0.4 \times 10^4 \text{ n/cm}^2\text{/sec}$$

As an example, assume that 10^{-4} of the neutrons will be in the energy interval 50 ± 5 kev. These neutrons will be detected with a counter having a cross section of 100 cm^2, a length of 20 cm, and filled to 1 atmosphere with $B^{10}F_3$; the (n,α) cross section of B^{10} is 2.5 barns at 50 kev. The counter efficiency turns out to be 1.3×10^{-3}, and the counting rate for the channel at 50 kev will be about 3 cpm which, while not overwhelming, would seem to be within the realm of possibility.

Neutrons produced in the (γ,n) reactions have energies of the order of 1 Mev and will probably have to be slowed down in paraffin. As can be seen from the table, the slowing-down times are quite short, so that this factor itself should not introduce a very great spread in the pulse. It should be noted that this situation is not quite the same as that for obtaining thermal neutrons. In the latter case the diffusion time in paraffin is quite long and a serious time spread in the source pulse is introduced.

It would be desirable to get along without having to pass the neutrons through a paraffin moderator. This could be done, however, only if a sufficiently large fraction of the neutrons were originally emitted from 1–100 kev.

TABLE 4
Some Neutron Characteristics

Energy	*Velocity* ($m/\mu sec$)	*Wave length* ($A.U.$)	*Number of collisions with hydrogen to reduce energy from 2 Mev*	*Estimated slowing-down time* (μsec)
1 Mev	13.8	0.0003	1	0.005
100 kev	4.38	0.001	3	0.01
10 kev	1.38	0.003	5	0.03
1 kev	0.44	0.0091	8	0.06
100 ev	0.14	0.0286	10	0.2
1 ev	0.014	0.286	14	2

The energy spectrum of the emitted neutrons is difficult to predict in detail, but a very rough estimate can be made as follows. Assume that the emitted neutrons have a Maxwellian distribution (that is, they result from evaporation from an excited compound nucleus)

$$dN/dE = (E/T^2)e^{-E/T} \qquad (9)$$

where E is the energy of the emitted neutron and T is the nuclear temperature. The fraction of the neutrons emitted in the range 0–E_0 is

$$\Delta N = 1 - (1 + E_0/T)e^{-E_0/T}$$
$$= \tfrac{1}{2}(E_0/T)^2 \quad \text{for } E_0/T \ll 1 \qquad (10)$$

If T is assumed to be about 1.4 Mev, which is a reasonable value for the nuclear temperature, and if $E_0 = 100$ kev, then

$$\tfrac{1}{2}(E_0/T)^2 = 0.25\%$$

This is not a very large fraction of the emitted neutrons but it might be possible to utilize it directly. In this respect, targets of high A are most effective, not only because of the larger neutron yields, but also because of the lower nuclear temperature (higher density of levels in the product nucleus).

Conclusions

It can be seen from the above that high-energy linear electron accelerators can be utilized to produce pulsed neutron sources for time-of-flight neutron velocity selectors. Preliminary, exploratory measurements with the 16-Mev accelerator at MIT, indicate that the neutron yields are as anticipated. Further improvements in this and other accelerators could result in pulsed neutron sources of intensity sufficient to permit the extension of the range of time-of-flight velocity selection to about 50 kev, thus bridging the gap between existing velocity selectors and monochrometers based on endoergic (p,n) reactions such as the Li(p,n) reaction. Perhaps the most important outstanding need is the development of a neutron detector of high efficiency in the 1–100 kev energy range.

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Nomogram for Estimating Decay of I^{131} and P^{32}

A rapid estimate, useful in routine work, of the present activity of either of these isotopes can be made with this nomogram if the activity at some previous time is known

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THREE METHODS exist for computing the activity of an isotope sample after a period of decay: 1. The present activity can be calculated by equation. 2. The per cent decay can be obtained from an exponential curve previously plotted. Multiplication of the past activity by the per cent decay will give the present activity. 3. A nomogram can be constructed to give the present activity directly.

While use of the decay equation is the most accurate method, it is time consuming, especially when isotopes are used routinely. A fairly accurate estimate of activity can be obtained with either of the other methods, but the nomogram on page 59 requires only one setting of a straight edge to provide an answer to problems involving I^{131} or P^{32} and decay periods shorter than a few half-lives.

Scale *A* represents the original activity as measured or calculated at a previous time, and scale *C* is the present activity. The time elapsed since the original activity was calculated is given, in days, on scales *B* and *B'*, for I^{131} on the right and P^{32} on the left. The scales of *B'* extend the range of the nomogram and are used for calculations that would not fall on the chart if the scales of *B* were used. When *B'* is used, the value obtained for the present activity has one less figure before the decimal point than the value of the original activity. When *B* is used, the answer contains the same number of figures before the decimal point. Sample problems indicated by dotted lines on the nomogram illustrate use of the two time scales.

Problem. A solution of I^{131} had a specific activity of 12.6 mc/ml. What is the specific activity 10 days later?

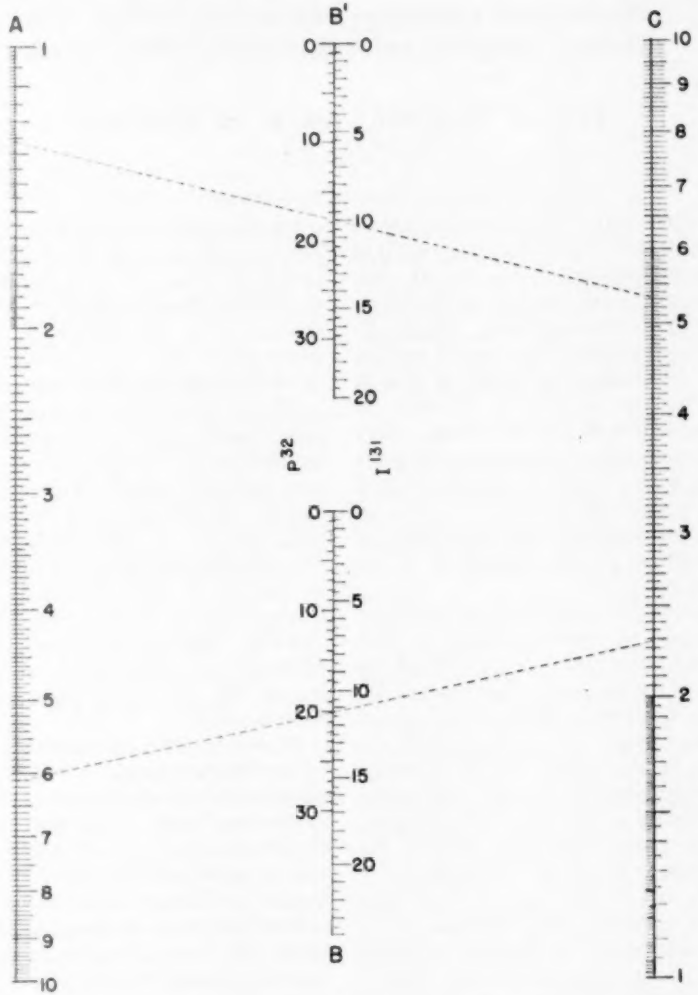
Solution. Place a straight edge on 126 on scale *A* and intersect the I^{131} scale of *B'* at 10. Read 532 on scale *C*. Since *B'* was used, the present activity must have a value containing one less figure before the decimal point than the original value of 12.6 mc/ml. The answer is therefore 5.32 mc/ml.

Problem. A shipment of P^{32} gives a dose rate of 6.11 mr/hr one foot from the surface of the container. What is the radiation at the same distance 20 days later?

Solution. Place the straight edge on 611 on scale *A* and intersect the P^{32} scale of *B* at 20. Read 229 on *C*. Since scale *B* was used, the answer is of the same order as the original activity, and the answer is 2.29 mr/hr at one foot.

Activity at some past time can be calculated from the present activity by working backwards. The use of scale *B'* then indicates that the answer will contain one more figure before the decimal point than the present value of the activity.

Nomogram for Estimating Decay of I^{131} and P^{32}



Instrumentation and Control of Reactors*

A myriad number of factors affect reactor control. The most important—reactor kinetics, space relation of control rods, load and temperature effects—are discussed in this summary. Instrument and mathematical problems are cited

By J. D. TRIMMER† and W. H. JORDAN‡

THE TWIN OBJECTIVES of reactor control are to permit operation of the reactor to fulfill its function, and to provide adequate protection against the radiation hazards of such operation. The principal danger which the control system must be designed to combat is that of allowing the reactor to develop an excessively high neutron flux. High flux would cause overheating of reactor parts, with consequent melting or similar damage to the reactor itself and with subsequent release and spreading of radioactive poisons dangerous to all living organisms.

Instruments and safety devices must be made to establish limits of flux beyond which the reactor will not be allowed to operate. The margin by which these safety limits should exceed the operating level is typical of the many factors making up the problem.

The purpose of the reactor determines the control problem to a large degree. In an isotope-producing reactor, the problem is to achieve maximum flux over a maximum volume, without anywhere exceeding safe thermal limits; time-dependence is relatively unimportant. In a research reactor, a given experiment may demand that at a particular location an accurately known flux be held precisely constant over a period of time. For a power reactor, the needs of the power plant should have dominion over the reactor; for mobile power plants this may make very strong demands on reactor time-dependence.

Efforts to develop mobile nuclear power plants, and other considerations tending toward higher neutron flux, have had an important effect on control design. Accumulation of fission products, which can affect stability and other dynamic properties of the reactor, is typical of nuclear factors which change markedly as flux levels of operation rise. There is also a general magnification of thermal problems, centering on the time-dependence of large heat transfers.

The scope of reactor control is shown in the block diagram of Fig. 1. The arrowheads indicate direction of flow of information and of energy. Broken lines indicate a relatively high proportion of information to energy. There appear in this figure three closed loops, around which closed-cycle actions can occur—the operator loop, the automatic loop and the load loop.

To discuss reactor controls, it is convenient to consider the parts of the control system separately. There can be no attempt to survey the entire subject of reactor instrumentation here, but this has been done previously (*1–5*).

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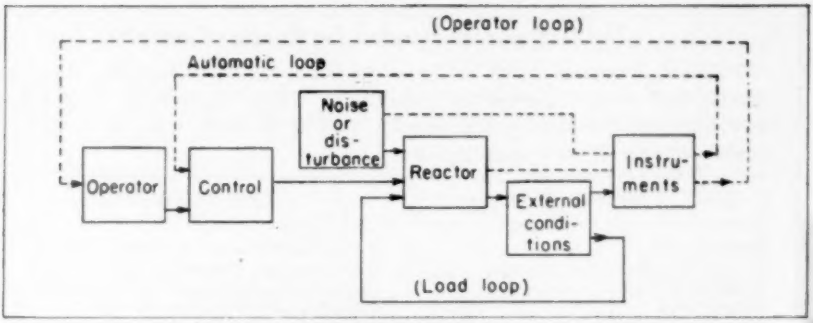

FIG. 1. General block diagram of reactor control

Several illustrative instrumentation problems are cited on page 62, and the rest of this article will be devoted to discussions of the main factors involved in control.

Effect of Reactor Kinetics

Some properties of the reactor itself affect control and must be considered in any discussion of control. In a thermal reactor, time-dependence of average lumped neutron density n may be described by a set of $m + 1$ first-order ordinary differential equations

$$\frac{dn}{dt} = A(t)n + \sum_{i=1}^{m} \lambda_i c_i + S(t) \quad (1)$$

$$\frac{dc_i}{dt} = g_i n - \lambda_i c_i \quad (2)$$

The m variables c_i represent density of sources* of delayed neutrons, $g_i n$ representing the rate at which the ith type of source is formed by neutrons being absorbed, and $\lambda_i c_i$ the rate at which it decays (forming delayed neutrons in the decay process). $S(t)$ is equivalent density of a direct source of neutrons. $A(t)$ is the reactivity coefficient, governing the multiplicative growth of neutrons, apart from the delayed-neutron effects.*

Consider now the solution of Eqs. 1 and 2 under equilibrium conditions (all derivatives zero) and with a constant source S_0 and with constant reactivity A_0. It is clear then, that

$$n = -S_0/(A_0 + \Sigma g_i) \quad (3)$$

Since it is possible to make the denominator of this expression equal to an arbitrarily small negative number, the possibility exists of multiplying a weak source to obtain a very large neutron density. From this it might appear that the task of reactor control would be to set and hold the denominator in Eq. 3 constantly equal to the appropriate "small negative number" to give the desired value of n. While this is not a bad first guess, it falls short on at least two counts: First, an impossible degree of regulation would be required, since the denominator might well be -10^{-9} so that regulation of one part in a million would still leave the reactor with thousand-fold ups and downs. Second, even if this kind of control were possible, it would be intolerably sluggish in changing from one desired condition to another.

So practical control of reactors calls for a wider variation of $A(t)$. Equation 3 shows that if A_0 exceeds some

* The appropriate number of source-types, i.e., the appropriate integral value of m, is open to some consideration. In the usual meaning of delayed neutrons coming from decay of fission products, it is customary to put $m = 5$ (*8*), although Kunstadter *et al.* (*9*) have reported additional weaker groups. In reactors containing materials such as beryllium and heavy water, there are additional sources from (γ,n) reactions (*10*).

* For derivation and integration of these Eqs. 1 and 2 see reference *11*.

The Instruments Needed for Reactor Control

To control a reactor, instruments are needed to furnish information about the reactor and about external load conditions. Both nuclear and nonnuclear quantities are measurable and can be used to indicate how the reactor is operating; among these quantities are:

Nuclear quantities	*Nonnuclear quantities*
Neutron flux	Temperature in reactor and coolant
Period of neutron flux change	Flow rate (of coolant or fuel)
Neutron energy spectrum	Liquid or gas pressure (of coolant or fuel)
Fission rate	Liquid level (of coolant or fuel)
Gamma radiation	Power
Reactivity (excess multiplication)	Motion and position of control elements
Fuel concentration (in liquid solutions)	

Not all the instruments and methods used to measure these quantities can be discussed here, but a few problems can be cited to illustrate typical control problems and their solution.

There is, for instance, the problem of knowing the reactivity, which is equivalent, as explained in the text, to knowing the reactor period. The early approach to this was to infer reactivity from observed reactor time dependence, but the delay involved is intolerable for short-period reactors. A much better solution now exists in the period meter. This meter is based on the logarithmic voltage-current characteristic of thermionic diodes (*6*, *7*), which converts a neutron signal n to a logarithmic scale log n. A resistor-capacitor differentiating circuit then gives $d(\log n)/dt$, which is proportional to the reciprocal of the period. The log n signal itself is extremely valuable for its wide-range indication of neutron level.

Another interesting problem is the separation of neutron signal from the intense radioactivity, particularly gamma rays, associated with the fission process and its products. Two methods, pulse discrimination and current com-

critical value A_c, so as to make the denominator positive, there would be no real (i.e., positive) equilibrium value of n. In this case the multiplication of neutrons would become infinite, and the presence in the reactor of even the tiniest source would ultimately lead to disaster. This makes it appear that it would be unsafe ever to take $A(t)$ beyond the critical value A_c, but everyone associated with reactors knows that they are commonly carried past criticality for limited times. To understand what actually happens, it is important to keep three points clearly in mind. The first is that every reactor contains some source, so that it is never strictly correct to speak of the zero-source form of Eq. 1. The second point, following from the first, is that a reactor, to be held indefinitely at a constant value of neutron flux, would have to be subcritical, i.e., $A < A_c$. The third point is that operating flux is generally very large compared to source strength, so that at such levels the "virtual sources" of delayed neutrons completely overshadow the true sources as far as transient effects are concerned.

Consequently one may draw the practical picture of reactor control as follows. At very low levels of flux, as in

pensation, are used. In proportional counters (such as those filled with BF_3) absorption of a neutron causes an event giving a stronger pulse of current than that caused by a gamma ray. By counting only the large pulses, the gamma rays can be ignored. Still better discrimination is obtained with fission chambers, in which uranium nuclei absorb the neutrons, releasing heavily-ionizing fission products.

Pulse-counting methods are especially useful at low neutron levels. The alternative method of current compensation is better at high levels, although it only works over a limited range of about 100 to 1. Ionization chambers having two active volumes, one of which is sensitive to neutrons and to gamma rays, the other to gamma rays only, yield a net current which, with proper relative size of the two volumes, is approximately independent of gamma-ray intensity.

Another neutron measuring instrument of great usefulness in reactors is the boron thermopile, in which the heat of neutron-induced radioactivity affects thermocouple junctions. These instruments are simple and compact, and in applications where their slower response is acceptable, they can serve instead of ionization chambers.

Besides instruments *per se*, circuits are required for combining information from a number of instruments. At low flux levels, where the hazard that short periods can create is important, it is reasonable to take safety action on the signal from one instrument, and in particular from the first instrument to call for action. Circuits have been developed to accept signals from several instruments and to initiate safety action in response to whichever instrument first puts out its signal beyond a certain level. These circuits were called "sigma circuits" (sigma for summation), but have later been more accurately dubbed "auctioneer circuits." At the high flux near operating level, the awkwardness of an unnecessary shutdown caused by defective instruments is greater and the danger involved in waiting for agreement from a quorum of instruments is less. It is therefore reasonable to shift from the auctioneer circuit to a coincidence circuit which initiates safety action only upon agreement of several (e.g., two out of three) of the instruments connected in the circuit.

starting up a new reactor, great attention is paid to the source strength and to its multiplication according to Eq. 3. The condition of criticality is cautiously approached from the sub-critical side. At higher levels the source may be ignored and reactor kinetics considered in terms of the zero-source form of Eq. 1, since the error thus incurred is no greater than is involved in other assumptions underlying these equations. Most control problems relate to the higher levels, and it is therefore reasonable to take the source-free form of the equations as a working basis. But it is important to know the significance of true sources, especially for start-up problems.

Zero-source kinetics may be visualized by considering typical responses to reactivity changes. Figure 2 shows the change of flux when the reactor has been at equilibrium with $n = n_0$ and with $A = A_c$, and the reactivity suddenly changes to a (positive or negative) constant value corresponding to $A = A_0$ at time zero. (The quantity l is the mean lifetime of the neutrons, in seconds.) Since by definition $A_c = -\Sigma g_i$, if we define $a = A_0 - A_c$, then $A_0 = -\Sigma g_i + a$ and Eq. 1 for time after zero becomes

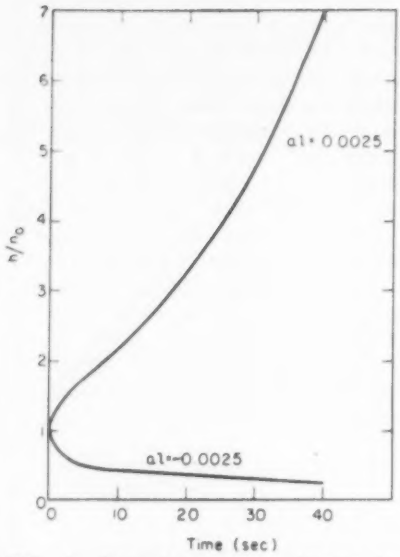

FIG. 2. Response to step changes in reactivity

$$\frac{dn}{dt} = an - \sum (g_i n - \lambda_i c_i) \qquad (4)$$

or, by Eq. 2

$$\frac{dn}{dt} = an - \sum \frac{dc_i}{dt} \qquad (5)$$

This helps in understanding the shape of the curves in Fig. 2. Immediately after zero, $dn/dt = an$ and n climbs (or drops) steeply. Then, as n increases, the dc_i/dt follow according to Eq. 2. This reacts back on dn/dt, as in Eq. 5, slowing down the neutron rise. Eventually the rise is a simple exponential, with time constant or period T given by the positive root of

$$T = \frac{1}{A_0 + \sum \frac{\lambda_i g_i T}{1 + \lambda_i T}} \qquad (6)$$

Figure 3 shows this relation in terms of T against a. Although it is common to single out the case of $a = \Sigma g_i$, this "prompt criticality" is merely a convenient landmark; the response in this case is qualitatively indistinguishable from that for neighboring values.

Another type of response of particular interest in control problems is that which occurs when A is a linear function of time. This corresponds to constant motion of a control rod of constant sensitivity. A typical curve is shown in Fig. 4.

The computation of curves showing response to arbitrary changes in A as do those in Figs. 3 and 4, is difficult. It is worth noting that the converse problem, of finding the time dependence of A that must have caused an arbitrary change in n, is much easier. Writing Eq. 2 as

$$\frac{dc_i}{dt} + \lambda_i c_i = g_i n \qquad (7)$$

it is seen that, if $n(t)$ is known, these are simply first-order equations with constant coefficients. With the c_i known from the integration of these equations, they can be put into Eq. 1 which may then be solved for $A(t)$ in terms of known quantities, and thus

$$A(t) = \frac{1}{n}\left(\frac{dn}{dt} - \sum \lambda_i c_i - S\right) \qquad (8)$$

The simplified discussion of reactor kinetics thus far presented must be supplemented, in many control problems, by consideration of how fission products affect reactivity. Suppose,

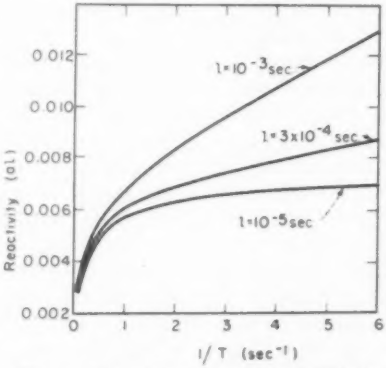

FIG. 3. Relation of period to reactivity

for example, that a given product P_f is radioactive and decays with rate $\lambda_f P_f$ into an element P_a with a large neutron absorption cross section, and with radioactive decay rate $\lambda_a P_a$. If the reactor has been operating for some time at a high level of flux, it will contain large equilibrium amounts of P_f and of P_a. These equilibrium values will be fixed by balances between the radioactive processes and the flux-dependent processes of fission and neutron absorption. If the reactor is shut down, so that the flux drops essentially to zero, this balance is upset and the poison concentration will increase and go through a maximum sometime later, as shown in Fig. 5 for two different values of pre-shutdown flux.

Besides having important practical bearing on control, this kind of effect is a serious mathematical complication, far more so than that due to the superficially similar effect of delayed neutrons. The big difference is due to the fact that delayed neutron groups simply add to the number of equations, whereas poisons do this and in addition make the system of equations nonlinear. The equations for P_f and P_a are

$$\frac{dP_f}{dt} = -\lambda_f P_f + g_f n \qquad (9)$$

$$\frac{dP_a}{dt} = -\lambda_a P_a + \lambda_f P_f - \sigma_a P_a n \qquad (10)$$

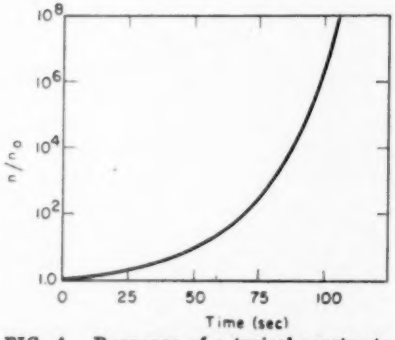

FIG. 4. Response of a typical reactor to constant reactivity increase of 5×10^{-3} per second (from reference 11)

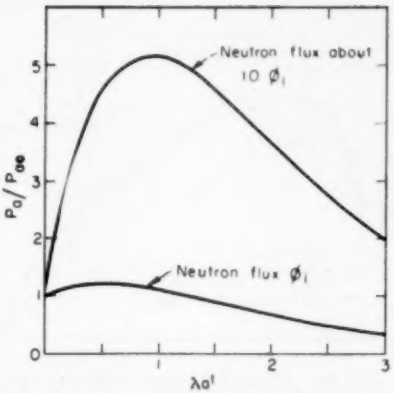

FIG. 5. Post-shutdown time-dependence of poison

These equations, with Eqs. 1 and 2, would constitute a numerous but still linear system except for the last term in Eq. 10 and the all-important fact that the coefficient A in Eq. 1 is now a function of P_a as well as of time; the system of equations is nonlinear and difficult to manage analytically.

Control-Rod Space Dependence

In the preceding discussion of reactor kinetics, attention was directed to the coefficient $A(t)$ as the vehicle of control. This quantity depends more or less on everything in and around the reactor, but for control purposes an attempt is made to localize the predominant effect in one or more control elements, the classic prototype of which is a poison-bearing rod. The effect of these control elements may be approximated, as in the preceding discussion, by treating the whole reactor as a single lumped system, ignoring all space-dependent variations. There are, however, important aspects of control which involve space-dependence.

Figure 6 shows the space-dependence of flux due to insertion of a single rod along the axis of a cylindrical reactor. This is evidently a rather drastic effect. The degree of insertion of one rod can

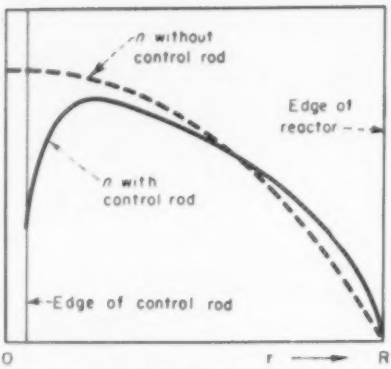

FIG. 6. Space-dependent effect of control rod

clearly have a strong effect on the total capacity as well as on the differential sensitivity of a neighboring rod, since both depend on the flux distribution in which the rod is working. It is usually good practice to keep the various control rods approximately in phase—that is, to avoid unsymmetrical configurations of some fully-inserted and other fully-withdrawn rods. One reason (not having to do with space-dependence) is that a fully-inserted rod is worthless as a safety element. The space-dependent consideration is that symmetrical configurations are less likely to entail undue thermal stresses and are more efficient in giving maximum total flux.

It is general practice to employ most of the control rods on a given reactor as slow, coarse controls (often called "shim rods") and to utilize one or two as fast, fine regulators. The regulating rod is usually in an automatic control loop. The sensitivity of the regulating rod's control action may be changed considerably by the gradual shift in position of the shim rods. This adds a rather nasty complication to design of the regulating-rod servo. Somewhat similar complications are inherent in the effect of control-rod location relative to measuring instruments, particularly those which inform the servo controlling the rod in question.

The Reactor and Its Load

The load loop shown in the block diagram, Fig. 1, is receiving increasing attention in reactor-control design, particularly for power-plant reactors. The effect of the load on the reactor is similar to that of control rods, in that it involves the coefficient $A(t)$, but it is thermal in action and so is different from the usual neutron-absorbing control element. Temperature changes in a reactor affect its reactivity in two principal ways which may be identified as nuclear temperature effect and density temperature effect.

The energy distribution of the neutrons is a function of the temperature, and absorption and scattering cross sections of nuclei for neutrons in turn depend on neutron energy. Thus the fuel, moderator, reflector, fission products—any or all may have cross sections sensitive to shifts in mean neutron energy and therefore to ambient temperature inside the reactor.

In addition to this nuclear temperature dependence, there is the density temperature dependence due to expansion of reactor materials. The entire reactor may increase in size and thus become more reactive. Also, mean free paths of neutrons inside the reactor will be changed by shifts in spacing between absorbing and scattering nuclei; in reactors with liquid coolant or liquid fuel, the expansion of these materials may cause additional effects.

Generally speaking, density temperature coefficients are negative—that is, reactivity decreases with increasing temperature. Nuclear temperature coefficients may be either positive or negative. The net temperature response of the total reactor is thus seen to be a balance of many factors. Magnitudes of individual factors vary greatly and hence the net reactivity allotted to temperature for the reactor as a whole depends very much on reactor design. The effects are large,

and an allottment of several per cent of reactivity is not uncommon.

For control purposes it is important to know the time constants as well as the magnitudes of these thermal effects. This is difficult, particularly in heterogeneous reactors. Consider a lump of fuel. The neutrons, which cause the heat-producing fissions inside the lump, enter into it freely, but the heat produced by the fissions may escape from the fuel element into the moderator or coolant comparatively slowly. More time must pass before the ambient temperature is shifted and the neutron energy affected thereby. Rates of heat transfer are highly dependent on materials, geometries, and flow rates. Hence thermal time constants vary widely from one reactor design to another, and from one portion to another of a given reactor.

The task of control design for maximum reliability and safety is materially simplified if the net reactivity temperature coefficient is negative. This self-regulating tendency is particularly valuable if it inheres in the fast temperature response—say, in the fuel, which is first to feel a rise in neutron flux in the reactor.

Consider now the load loop of a reactor through which coolant is pumped to a heat exchanger (see Fig. 7 for a block diagram). Changes in temperature T_i of coolant entering the reactor will affect the reactivity coefficient, $A(t)$, which in turn will affect neutron flux and rate of heat-producing fission. The outlet temperature T_0 of coolant leaving the reactor is thus changed, and in passing through the heat exchanger is brought down again to T_i.

Each of the four links in this chain of events may be approximately described by an equation or functional relationship. The effect of temperature on reactivity is so complex that no one generalization is worthwhile. For small excursions a linear relationship,

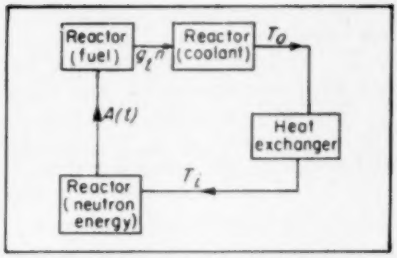

FIG. 7. Block diagram of thermal load loop

based on a constant over-all temperature coefficient, is reasonable.

Effect of reactivity on neutron flux has already been described by Eqs. 1 and 2. This leaves the heat exchange from reactor to coolant and from coolant to heat exchanger. Both of these may be approximated by simple first-order equations. In the reactor let $g_l n$ be the rate of heat production due to fission, C_f the heat capacitance of the fuel, and R_f the thermal resistance between fuel and coolant. Then T_f, the fuel temperature, is related to the mean coolant temperature T_m by the equation

$$R_f C_f \frac{dT_f}{dt} + T_f = T_m + R_f g_l n \quad (11)$$

The rate at which heat is conveyed to the coolant is $C_c(T_0 - T_i)$ where C_c represents heat capacity of coolant flow. This must satisfy the equation

$$C_c(T_0 - T_i) = \frac{T_f - T_m}{R_f} \quad (12)$$

Similarly, in the heat exchanger

$$C_c(T_0 - T_i) = \frac{T_m - T_e}{R_e} \quad (13)$$

where R_e is the thermal resistance and T_e the mean secondary temperature in the exchanger.

In writing down these simple equations, many crude approximations are involved (among others, neglect of transport lags between reactor and heat exchanger). The purpose in writing them is to point out that even with simple equations there are still two great difficulties. One is determination

of constants entering such equations—represented by R's and C's in Eqs. 11 to 13. The other difficulty is in the relation of these equations to Eqs. 1 and 2. Since the reactivity coefficient A will be a function of T_f, T_m, etc., as well as of time, the complete system of equations is nonlinear, just as in the case of fission-product effect.

Mathematical Status of Control

Whenever a system is controlled through a coefficient of its differential equation—exemplified here by $A(t)$ in Eq. 1—the over-all equation for the closed control loop becomes nonlinear. This is inherent in the definition of closed-cycle control, which means that the control action is made to depend on the output quantity—that is, the dependent variable of the system's equation. If the control action is by direct forcing (exemplified by the source S in Eq. 1) rather than by forcing of a coefficient, the complete control equation remains linear. But when a coefficient becomes a function of the dependent variable, the equation is necessarily nonlinear. Thus , closed-loop control is a third source of nonlinearity in the reactor control equations, in addition to the two mentioned earlier—fission-product and temperature effect on reactivity.

In an attempt to apply the familiar transfer-function techniques appropriate to direct-forcing problems to the coefficient forcing of reactors, one can proceed by investigating first the phenomena of small variations about an equilibrium situation. This reduces both the open-loop and the closed-loop problems to one of constant coefficients, synonymous with direct forcing, in a linear equation. Thus if one puts $A = A_0 + A'$ and $n = n_0 + n'$ in Eq. 1, with the understanding that the time-dependent primed quantities are so small that their powers and products may be neglected, it becomes

$$n_0 \frac{dn'}{dt} = A_0 n' + \sum \lambda_i c_i + n_0 A' + S + A_0 n_0 \quad (14)$$

This is a linear equation with constant coefficients, to which the usual transfer-function techniques and terminology are applicable. But it should be noted that the process has meaning only for small variations. The over-all study of the reactor control equations from the general viewpoints of nonlinear mechanics should not be neglected.

A powerful tool for studying nonlinear problems is the use of analogs or simulators. These are very helpful in the control-loop and fission-product aspects of reactor nonlinearity, where specific data is available to establish simulator parameters. Comparable success might be obtained in the thermal problems, were it not for the general intransigeance of heat-transfer phenomena. Heat-transfer physics is difficult to analyze, difficult to scale down for study on models, difficult to measure on full-scale equipment. It is is difficult, period. The lack of quantitative understanding of thermal, thermo-chemical, and thermo-physical phenomena is the greatest obstacle in the path of developing valid reactor analogs.

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BOOKS

The Chemistry of Uranium (Part I)—The Element, Its Binary and Related Compounds (Div. VIII, Vol. 5, of National Nuclear Energy Series), by Joseph J. Katz and Eugene Rabinowitch, McGraw-Hill Book Co., Inc., New York, 1951, xxi + 609 pages, $7.25.*

This book, which is concerned with the "dry chemistry" of uranium, is to be followed by a companion book devoted largely to the properties of ionic uranium in aqueous solution. In anticipation of the completion of the treatise with the pattern set by the first book, one feels free to predict that the volume will remain for many years the definitive work on uranium chemistry.

The book at hand comprises a critical as well as comprehensive synthesis of the subject and goes beyond a mere mechanical recording of reported data. In this respect, it represents a different kind of effort from the "Uranium Volume" of the Gmelin series (published in 1936). It incorporates the voluminous wartime and postwar work associated with the atomic-energy programs much of which, it can be inferred, is being presented here in the open literature for the first time. A rough estimation indicates that the wartime output has more than doubled the literature on this subject. All told, this volume clearly represents a prodigious amount of discriminating effort.

This reviewer found the candid style of presentation employed by the authors highly refreshing in a technical work. Throughout, one feels clear on the author's thoughts, both of affirmation and doubt. Where conflicting data have appeared, the authors endeavor to make selections; where none can be made, this status is clearly pointed out.

Another general feature of the book worthy of special comment concerns the types of information incorporated. Along with the recording and evaluation of fundamental properties such as thermodynamic constants, the authors also present a comprehensive coverage of less quantitative descriptive matter. Although this feature necessarily obviates a short treatment and may impair its usefulness to the student or casual reader interested only in basic properties, its value to the research worker is overwhelming.

The detailed information present in this book is reduced to size by the use of frequent delineating headings arranged in outline form. The subdivisions under which uranium trichloride is covered in eleven pages in the chapter on *Uranium-Chlorine Compounds* is given here as an example:

1. Uranium Trichloride

1.1. *Preparation of uranium trichloride*
- (*a*) UCl_4 + hydrogen
- (*b*) UH_3 + HCl
- (*c*) Miscellaneous preparations
- (*d*) Purification of UCl_3

1.2 *Physical properties of uranium trichloride*
- (*a*) Melting point
- (*b*) Volatility
- (*c*) Crystal structure of UCl_3
- (*d*) Thermochemical data

1.3 *Chemical properties of uranium trichloride*
- (*a*) Water
- (*b*) Other solvents
- (*c*) Thermal decomposition of UCl_3
- (*d*) Oxidizing agents
- (*e*) Reducing agents
- (*f*) Complex compounds of UCl_3

The index is fairly brief but will probably not receive much use in any case because of the ease of locating subject matter after some familiarity with the format is attained. Reference indices are given throughout the text with

* EDITOR'S NOTE: Although this book carries the notation "Part 1," the information in it is complete and is not to be construed as introductory or accessory to the material planned for inclusion in the second half (Part 2) of the volume to be published later.

references listed after each chapter. A rough estimation would place the number of references at greater than 2,000.

The book begins with a short chapter titled *Isotopic Composition and Atomic Weight of Natural Uranium*. This is followed by a detailed chapter on the X-ray and optical spectra which contains a discussion of the electron configuration. The third chapter, *Uranium in Nature*, gives a digest of the geochemistry of uranium and includes a helpful classification of uranium minerals.

The following four chapters pertain particularly to uranium metal: the extraction of uranium from ores and metal preparation, physical properties, chemical properties, and intermetallic systems.

The remainder of the book (418 pages) is taken up with chapters devoted to a systematic survey principally of the binary compounds of uranium. That the literature on some of the subjects has become voluminous is evidenced by the fact that 84 pages are devoted to oxide systems of uranium and 53 pages to the single compound UF_6. To illustrate further the inclusion of information useful to a research worker may be cited a section dealing with the handling of UF_6.

As may be inferred from preceding statements, this book is one to excite a great deal of enthusiasm. It is well organized, written with clarity, and extensive use is made of tables and graphs. The treatment of chemical and physical properties is modern, and good evidence is given that data old and new have been re-evaluated and brought into perspective. Inorganic chemists would benefit greatly by the availability of reference books with equally competent treatment of other elements.

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BOOKS RECEIVED

After the A Bomb? (Emergency Care in Atomic Warfare), edited by Charles F. Behrens, M.D., Adm., MC, USN, Thomas Nelson & Sons, New York, 1951, ix + 182 pages, $2.50. Eight medical men, in addition to Admiral Behrens, have each contributed a chapter to this practical and intelligent guide to health problems and medical care following an atomic attack. It goes into details that every civil defense unit in the country should be prepared to consider.

Herbert H. Dow, Pioneer in Creative Chemistry, by Murray Campbell and Harrison Hatton, Appleton-Century-Crofts, Inc., New York, 1951, xi + 168 pages, $3.50. A biography of the founder of the Dow Chemical Company.

Fundamentals of Electronics, by F. H. Mitchell, Addison-Wesley Press, Inc., Cambridge, Mass., 1951, xi + 243 pages, $4.50. A college text, with problems and answers. Instrumentation is stressed.

200 Miles Up—The Conquest of the Upper Air, by J. Gordon Vaeth, The Ronald Press Company, New York, 1951, xiii + 207 pages, $4.50. Source of information on atmospheric research, rocketry, and ballooning. The chapters on *Upper Air Research Instruments*, *Vehicles for Upper Air Research*, and *Project Skyhook—Upper Air Research by Balloon* will be of general interest to cosmic-ray researchers.

Radioactivity Applied to Chemistry, edited by Arthur C. Wahl and Norman A. Bonner, John Wiley & Sons, Inc., New York, 1951, xv + 604 pages, $7.50. (*To be reviewed.*)

Preparation, Properties, and Technology of Fluorine and Organic Fluoro Compounds (Div. VII, Vol. 1, of National Nuclear Energy Series), Charles

Slesser, editor, Stuart R. Schram, associate editor, McGraw-Hill Book Co., Inc., New York, 1951, xxiii + 868 pages, $10.50. *(To be reviewed.)*

OTHER LITERATURE

Principles of Civil Defense Operations (Web Defense, Mutual Aid, Mobile Support) (AG-8-1, 45 pages, $0.20); **Water Supplies for Wartime Fire Fighting** (TM-9-1, 14 pages, $0.10); **The Warden Service** (AG-7-1, 44 pages, $0.20); and **Civil Defense in Industry and Institutions** (AG-16-1, 60 pages, $0.25). Four new pamphlets prepared by the Federal Civil Defense Administration. *GPO, Washington 25, D. C.*

Iodine, Its Properties and Technical Applications. Contained in this 74-page booklet is the story of the chemistry of iodine and its compounds and how they are used in organic chemistry, inorganic chemistry, metallurgy, and electrical and electronic applications. It is extensively referenced to the literature. A companion pamphlet, **Physical Properties of Iodine and Its Inorganic Compounds** (Iodine Abstracts and Reviews, Vol. 2, No. 1, Jan.–Apr., 1951) is a tabulation of over 500 substances, with information on density, melting and boiling points, and solubility. *Both available without cost from Chilean Iodine Educational Bureau, Inc., 120 Broadway, New York 5, N. Y.*

French Bibliographical Digest—Science, Physics (June, 1951). An 87-page booklet listing the most significant books on physics published in France between 1940 and 1948. *Available without cost from Cultural Division of the French Embassy, 934 Fifth Avenue, New York 21, N. Y.*

How to Be Cleared for Handling Classified Military Information Within Industry. Prepared by the Office of Industrial Security, Munitions Board, Dept. of Defense, in June, 1951, this 7-page folder covers information on both plant and employee clearance. *GPO, Washington, 25, D. C., $0.10.*

Industrial Waste Disposal—Radioactive Materials (Bulletin C-27). A brief description of sources and types of waste, their effects and disposal, and insurance coverages involved. *Available without cost from Association of Casualty and Surety Companies, 60 John Street, New York 38, N. Y.*

Next month in **NUCLEONICS**

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containing the answer to "Who makes it?" and "Where can I get it?" for over 500 products used in the field of nuclear science.

2. CIVIL DEFENSE SECTION . . .

containing hitherto unpublished data and definitive information for the nuclear scientist active in the technical aspects of civil defense planning.

CROSS SECTIONS

A Simple Radioactive-Thyroid Phantom

By L. E. Preuss
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Research and clinical laboratories using I^{131} in human and animal studies quickly recognize the routine need for a phantom of the thyroid and surrounding neck region. Such a device is necessary both in routine and research measurements involving metabolic and dosage studies. Certain necessary requirements exist for such a device. It must be made of easily procurable materials, be disposable, simply set up, light in weight, inexpensive, of negligible health-physics hazard, and it must be as close a duplication as practical of the *in vivo* phenomena of geometry, size, shape, self-absorption and backscatter.

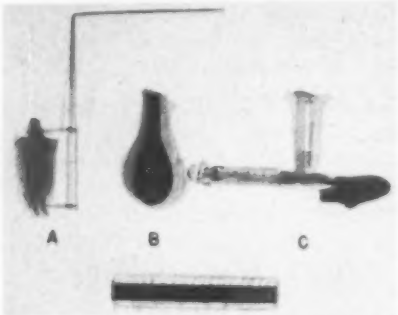

FIG. 1. A is the glove finger supported in the wire frame. B is a toy balloon suitable for the phantom. C shows the method of injecting I^{131} solution into the glove finger

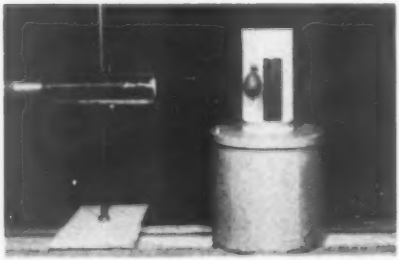

FIG. 2. Cut-away view of the mock thyroid and backscattering cylinder positioned in the carton. A shielded end-window G-M counter is positioned at left

Of the several phantoms in common use at present each has certain disadvantages. The most common thyroid phantom, consisting of a 25-ml volumetric flask immersed in water in a 1,000-ml beaker (*1*), introduces errors concurrent with differences in absorption, geometry, and backscatter of glass as compared to living tissue. Freedberg, *et al.* (*2*, *3*) and others (*4*) have discussed these I^{131} uptake measurement problems. The cadaver method (where the thyroid is injected with I^{131} or substituted for by a plastic gland containing I^{131}) is excellent for the reproduction of physical phenomena but obviously does not lend itself to routine work. The elaborate Lucite, Vinylite (*5*), and Plexiglas phantoms, filled with water or gelatin, and the laminated Masonite models are complicated in fabrication, are expensive, fail in health-physics requirements, and fit poorly into routine work. Wax and blown-glass phantoms have been described by Oddie and Scott (*6*). Swallowing of

phantoms has been described by Pochin (*7*).

An exceedingly simple and useful phantom has evolved from the needs of this laboratory for a mock-up of the human thyroid.

An ordinary spherical or ellipsoidal toy balloon of about 3 or 4 cm optimum diameter (B, Fig. 1) serves as the "thyroid." This size will accommodate 20 to 30 ml of I^{131} solution without expanding the balloon. (The normal human thyroid equals roughly 25 ml.) The balloon may be sealed off without pressure on the solution, and danger of leakage of the aliquot is thus reduced.

The fingers of ordinary surgical gloves (size 7 to 9) may also be used as "thyroids" when cut from the glove, sealed with rubber cement at the cut, and filled by means of 2 syringes with a 24-gage needle and a 3-way stopcock (C, Fig. 1). A solution of I^{131} may be introduced from the vertical syringe via the horizontal syringe into the "gland." This scheme will facilitate rinsing by means of the same technique. Two rubber tabs connected on each end are useful for attaching the finger to a support (A, Fig. 1). The two lobes of the thyroid may be imitated by joining two 10–15-ml glove fingers.

The single toy balloon or glove finger is satisfactory for the routine I^{131}_{4} percentage-uptake measurement taken at or beyond 22 cm (*8*) where these phantoms act much as a point source.

To approximate the geometry of the thyroid in the neck, the rubber "gland" is attached to a light wire framework and suspended in a 2-quart ice cream carton* as shown in Fig. 2. The pressed-paper carton, when filled with water, offers absorption and backscatter somewhat similar to the *in vivo* conditions. The carton may contain a dilute I^{131} solution to simulate the concentration of I^{131} in the blood and tissue (other than thyroid) of the neck.

Those cartons not waterproofed may simply be coated with a thin layer of paraffin by rotating the melted wax in the interior and pouring off the excess.

The wire framework device (A, Fig. 1) has proved useful for exact positioning of the gland in the carton. Numbers 14 and 20 gage tinned circuit wire are satisfactory for this frame.

To simulate backscatter produced by dense tissues and the bones of the spine, a solid plaster-of-paris cylinder is used (Fig. 2). Estimates of percentage increase of forward radiation due to scatter vary from 10 to 30% for I^{131} radiation from the adult human thyroid. The individual worker may study this phenomena *in vivo* (*9*) and then reproduce the desired range in the phantom by varying the atomic number of the backscatterer and its size and proximity to the source. To reproduce the physical effects of the chest and head volumes a 2½-gallon carton may simulate the chest while a 1-gallon carton may be used as a "head." The "neck" may be shortened to match the conditions found in the living subject. Scatter from the rib cage and skull is difficult to imitate.

The phantom technique as described, best applies to human adult studies. However, this same technique may be used on studies of infants and children. Here the geometry and size relationships differ greatly from the method outlined above. Since endless varieties of paper cartons are commercially available, suitable phantoms may be made for these special cases.

The balloons and rubber fingers should be discarded after a few days use since they become brittle with age. The used cartons may be incinerated.

In summary, the component parts of the phantom are routinely available as well as inexpensive. The cost for one set-up is well under one dollar. The

* These are available corresponding to about a size 15 male collar or about 4½ in. diameter and 8 in. high.

whole unit is easily assembled and light in weight. It produces a reasonably close duplication of the *in vivo* phenomena. The health physics problems are slight, and disposal is easily effected by incineration (*11*).

Because of the widely variant human subjects involved, even the most elaborate models only approximate the living situation (*8, 10*). This approximation may introduce large errors, a fact which applies to the methods outlined here.

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*A New Film Packet For Radiation Dosimetry**

By William C. Herman
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Type 552 film packet, widely utilized for personnel monitoring by various installations, only covers a range of exposures from approximately 50 mr through 50 r satisfactorily. The packet has a two-fold purpose when used for personnel monitoring: first, to record low exposure values such as are received by people operating within the prescribed exposure limits; second, to record high exposures such as might occur in the event of a serious accident. In addition, the 552 packet has been used to some extent for other problems of dosage measurement.

Disadvantages of 552 Packet

The gamma-ray response of the 552 packet is shown in Fig. 1. This packet contains two films, sensitive and insensitive, designated as type 502 and 510 respectively. One set of exposed films was processed in a standard X-ray developer, du Pont concentrated X-ray developer (curves 1 and 2); another set was processed in a fine-grain developer (curves 3 and 4).

A consideration of the calibration curves of the 552 packet reveals that it falls short of the ideal in several respects:

1. It doesn't go down to as small values of exposure as would be desirable for ordinary health physics use. Fifty mr is about as small a value of exposure as can be detected routinely with the sensitive film in 552 (i.e. 502), and this is the daily exposure limit at most laboratories.

2. The insensitive film does not go up nearly as high as would be desirable for use in emergencies, civil defense or various experimental applications.

3. As one can see from the curves in Figure 1, fine-grain development is no

* This work was done under the auspices of the U. S. Atomic Energy Commission.

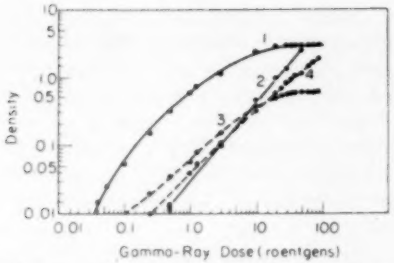

FIG. 1. Gamma-ray response of films in standard 552 packet. Curve 1, sensitive film (du Pont 502); curve 2, insensitive film (du Pont 510), regular development. Dotted curves 3 and 4, same films using fine-grain development

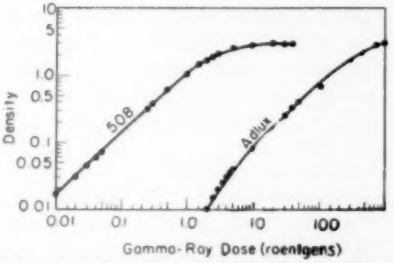

FIG. 2. Gamma-ray response of films proposed for new packet, with standard development

solution since it extends the upper limit of the insensitive film at the expense of low-dosage coverage by the sensitive film, although for special purposes where the really low doses are of no interest, this method can be of value.

Proposed New Film Packet

The ideal packet would have a sensitive film with a sensitivity as high as is compatible with good photographic characteristics and an insensitive film that would measure down to the upper limit of the sensitive and up to as high an upper limit as possible.

A study has been made of a number of standard emulsions with the thought of selecting a combination that would have more ideal characteristics. A very good combination is a packet containing du Pont type 508 X-ray film as the sensitive component, and Defender Adlux as the insensitive film. Calibration curves for these films, using du Pont concentrated liquid X-ray developer, are shown in Fig. 2. The following favorable characteristics may be pointed out:

1. The sensitive film can now be read down to about 15 mr as compared with 50 mr for type 502 at the same density.

2. The ranges of the two films overlap satisfactorily in the vicinity of 5 r.

3. The useful dosage range of the Adlux film extends to approximately 750 r.

Processing of all films used was done by the time-and-temperature method. Development in du Pont concentrated liquid X-ray developer was at 20° C for 3 minutes, with agitation every 15 seconds. Fine grain development was in Edwal Super 20 at 21.1° C for 12 minutes. After thorough washing, the films were rinsed in a wetting agent to eliminate water marks resulting from forced drying.

All film densities were read on a Photovolt photoelectric densitometer (Model 500) which has a density range from 0 to 4. Careful zeroing with a blank film was made regularly to keep densitometer errors at a minimum. A blank film processed with each set of exposed films was used to zero the densitometer. Density readings of all exposed films are net densities above emulsion and base fog. Three to four density readings were taken on both open-window and shield portions of the film, and average densities were used.

To replace the 552 packet for personnel monitoring and miscellaneous dosimetry applications, a new packet containing 508 and Adlux films would be a great improvement. The versatility of this film combination in its exposure coverage and wider range of application makes possible the standardization of one film packet for use by

many organizations requiring a film dosimeter. Through the courtesy of E. I. du Pont de Nemours and Co., a supply of such packets has been procured and will be tested in actual use.

Use Bottle-Crown Planchets In Radiotracer Studies

By Michael Marcour and J. T. Woolley

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Logan, Utah

For several months this laboratory has been using ordinary beverage bottle crowns (obtained from Western Crown Cork and Seal Corporation, 2650 Bay Shore Blvd., San Francisco, California) as planchets in radioiron, radiophosphorus, and radiozinc analyses of plant tissues.

One-half gram or less of dried and ground plant material is weighed into the crown and ashed at 550° C. Counting is done in a normal manner upon the ash in the crown.

Beverage-bottle crowns without cork inserts cost about 20 cents per gross. At this price crowns can be discarded after one use. The necessity for cleaning planchets and checking them for activity is thus eliminated.

Oil-Well Acidizing Controlled By Radioactivity Instruments

Radioisotopes now aid in increasing oil production by a new and simpler method of controlling and evaluating oil-well acidizing. The new method, developed jointly by Radioactive Products, Inc., Detroit, Mich., and Dowell, Inc., Tulsa, Okla., has been used routinely for several months in the oil fields of Kansas and Oklahoma. Dowell, a subsidiary of the Dow Chemical Co., is an oil-well servicing organization.

Acid is pumped into both new and old oil wells to dissolve particles of limestone that clog the pores of permeable oil-bearing formations and prevent the flow of oil into the bore-hole. Many new wells will produce no oil until such treatment has been given. Old wells are frequently revitalized by the process.

A method had been developed by Dowell for limiting the site of acid action to the oil-bearing formation only, preventing the wasteful reaction of acid on rock or limestone containing no oil. However, the pipe had to be removed from the oil well before the control instruments could be used. This involved the hoisting and complete disjointing of as much as 10,000 feet of pipe. The new radioisotope method of treatment eliminates this procedure.

Radioactive materials are processed and diluted to a standard strength at Dowell's Tulsa laboratories. These materials are then shipped to field stations where they are added to the oil-well treating acid.

Thousand-gallon tank trucks transport the radioactive acid to the oil well to be treated. An "electric pilot" truck containing Geiger counters and recording equipment is also sent to the well.

The Geiger counter used is 6 ft long and 1 in. in diameter. It was designed to withstand external pressures of 10,000 lb/in² and the effects of hydrochloric acid at the temperature of boiling water. It is lowered by means of a 12,000-ft steel and copper cable to the point in the well which is to be treated.

The radioactive acid is pumped down the 2-in. tubing that opens into the bottom of the well. The Geiger-counter instruments show a slight response as the acid streams past the counter. As more acid is pumped into the well, the level of the acid rises in the 6 to 10 in. annular space around the tubing. When this acid in contact

with the rock formation reaches the level of the counter, there is a sharp increase in the counting rate. This indicates that the acid has reached the desired level.

A second pump is then started to increase the pressure on the liquid in the annulus, forcing the acid into the porous formation. By adjusting the speed of the two pumps it becomes simple to move the level of the acid up or down and to hold it at the desired level during the treatment.

After acidizing is completed, water is flushed through the tubing to wash out the last traces of the radioactive acid. The Geiger counter is then lowered to the bottom of the well, and then raised while the radioactivity of the formation is recorded at each level. Thus it is possible to gain valuable information as to where the acid eventually went into the formation.

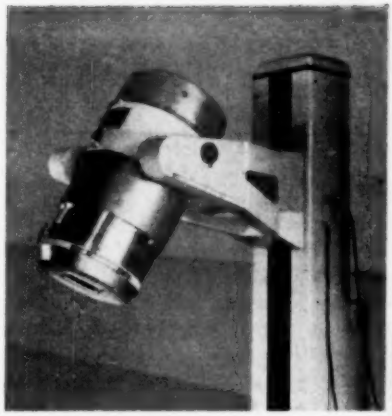
Isotope Therapy Unit Built For Use with 50 r/min Sources

The model A beam therapy unit for use with cobalt-60 sources has been made available commercially by the Eldorado Mining and Refining company of Ottawa, Canada. The unit incorporates sufficient protection for housing sources producing up to 1-Mev gamma radiation at intensities as high as 50 r/min at 1 meter.

Convenient controls on a flexible cable simplify angulation of beam within a 105 degree arc and height adjustment of the source between 28 and 81.5 in. from the floor.

A remote control panel is used to operate, time, and indicate the position of the shutter mechanism. This is a gravity-closing method of moving the shielding to form a conical port in the housing to expose the radiation beam; it does not involve moving the source.

A diaphragm system for adjustment of treatment field size and shape has



been provided. By a simple manual setting of two levers, square or rectangular apertures of infinite variety from 3×3 cm to 18.5×18.5 cm may be obtained. The apertures can be easily rotated completely about the beam axis.

The diaphragm is a single-plane type, and its distance from the source can be manually adjusted to suit requirements of the user since the lower section containing the beam-defining blocks can be telescoped into the upper portion for over 22 cm. A built-in optical system and light source reproduces the radiation beam, facilitating set-up.

Over-all height of the unit is less than 100 inches, and total weight is approximately 7,500 lb. This is the first commercial unit of this type to have been tested with a full-strength Co^{60} source; it will be installed for clinical use during the current month.

TECHNICAL BRIEFS

➢**Pulse-height analyzers** ordinarily have two discriminator levels and one recorder for each channel in the U. S., but the English have developed one discriminator level to distinguish all channels and one recorder for the group. Currently the British plan building one analyzer for 80 channels.

NUCLEONIC EVENTS

IRE NUCLEAR SCIENCE SYMPOSIUM PLANS ANNOUNCED

The tentative program for the Nuclear Science Symposium to be sponsored by the Institute of Radio Engineers, in cooperation with the Atomic Energy Commission, and held at Brookhaven National Laboratory, Upton, N. Y., from December 3–4, 1951, was released as follows last month by Urner Liddel of the Office of Naval Research, chairman of the program committee.

December 3: Beginning in the afternoon, the program includes papers on "Brookhaven Facilities" by L. J. Haworth, Director of Brookhaven National Laboratory; "Electronic Research at Brookhaven," by J. P. Blewett, BNL; "Nuclear Research of Engineering Interest," by D. J. Hughes, BNL; "The Secrecy Problem on Nuclear Engineering Information," by J. G. Beckerley, AEC; "Recent Developments in Nuclear Instrumentation in Health Physics," by H. M. Parker, Hanford Works (G.E.); and "The Problems of Liaison Involved in Conducting Information Services for Engineers," by N. H. Jacobson, AEC. An evening session will include a dinner, a talk on "Atomic Energy and Engineering," by T. Keith Glennan, member of the AEC, and a panel discussion on "The Role of Electronics Engineering in the AEC Developments," in which Commissioner Glennan, Keith Henney, editor of NUCLEONICS, and Clark Goodman, physics professor at Massachusetts Institute of Technology, will participate. L. V. Berkner, president of Associated Universities, Inc., will serve as moderator.

December 4: A morning session will include papers on "Applications of Scintillation Counters to High-energy Physics," by Robert Hofstadter, Stanford University; "Assessing the Function of Small Research Reactors," by Clifford K. Beck, North Carolina State College; "Some Recent Nuclear Developments at MIT," by Martin Deutsch, MIT; "Using Fission Products in Industry," by F. J. Lovewell, Stanford Research Institute; and "Industrial Interest in Nuclear Reactors," by P. N. Powers, Monsanto Chemical Co. A tour of research facilities at Brookhaven during the afternoon will conclude the meeting.

Arrangements for attendance can be made through Mrs. Mariette Kuper, Brookhaven National Laboratory, Upton, N. Y. Registration and dinner fee for the conference will be $5.

HANFORD TO BUILD LABS FOR METALLURGY, PILE TECHNOLOGY

Plans to construct a radiometallurgy building and a pile technology building for the Hanford Works, operated by the General Electric Company, in Richland, Wash., were revealed recently when construction bids were invited.

The radiometallurgy building will be one story high, with a full basement. Its main work area will be a long, narrow canyon in the center; supporting facilities will be located in wings on either side. It is expected that the new facility will be equipped for handling samples up to 1,000 curies activity.

The first floor will be reinforced concrete. It will be supported by concrete columns in the canyon and by steel beams and concrete columns in the wings. Interior walls will be of mova-

ble sectional metal partitions and liners.

The canyon, which will be 32 feet wide and 174 feet long, will have cells located along the longitudinal axis. Dry and wet sample storage facilities will be built into the west end of the canyon. A decontamination room and equipment-storage rooms are planned for one of the wings of the building.

The pile technology building will have two stories and a basement. It will be an L-shaped building, with outside dimensions of 180 and 190 feet.

The basement will have a concrete floor slab and reinforced concrete walls. The first and second floors will be steel frame with insulated steel panel outside walls and concrete floors. The basement partitions will be of gypsum block and reinforced concrete. Movable, sectional metal type partitions will be used on the first and second floors.

These are the last two units in the group planned for the Hanford Works Laboratory area, which is located about seven miles north of Richland. The other two units, the radiochemistry and health instruments control and development buildings, are already under construction.

NUCLEAR-POWERED PLANE, SUBMARINE CONTRACTS LET

Propulsion by nuclear power came nearer actuality recently with the awarding of contracts for the construction of a submarine and an airplane whose driving thrust would be derived from a nuclear reactor built into the craft.

The contracts included one let by the Navy on August 22 to the Electric Boat Co., Groton, Conn., for the design and construction of the hull of the atomic-powered submarine. Westinghouse Electric Corp., which has been working with the Atomic Energy Commission in designing and building an experimental nuclear reactor for naval vessels, revealed on August 30 that it had been awarded a Navy contract to provide the propulsion machinery for the submarine.

On September 5, the Air Force announced it had ordered the development of a suitable airframe for a nuclear-powered plane through a contract given to Consolidated Vultee Aircraft Corporation, San Diego, Calif. The General Electric Company is already at work on the nuclear-power plant for the aircraft.

ARCHITECTS CONFERENCE DATE CHANGED, SPEAKERS NAMED

According to revised plans announced last month, the Conference on Laboratory Design for Handling Radioactive Materials, originally scheduled for October (NU, Aug. '51, p. 79), will now be held from November 27 to 28. Jointly sponsored by the American Institute of Architects and the Atomic Energy Commission, the meetings will take place at the National Academy of Sciences in Washington, D. C.

The program, as it now stands, includes the following subjects and speakers: *Design of Laboratory Facilities for Research with Radioactive Materials at Various Levels*, Alexander Mackintosh; *Air Supply and Exhaust for Laboratories Handling Radioactive Materials*, Carleton P. Roberts; *Shielding for Various Levels of Radioactivity*, Nelson B. Garden; *Surfaces and Finishes for Radioactive Laboratories*, James G. Terrill; and *Waste Disposal*, E. C. Pitzer.

Each of the subjects will be discussed by a panel, then opened to a general discussion from the floor.

AEC EXTENDS G.E.'S HANFORD, KNOLLS CONTRACT

The Atomic Energy Commission last month announced it has renewed for five years the General Electric Company's contract for operation of the Hanford (Wash.) Works and the Knolls

Atomic Power Laboratory, Schenectady, N. Y.

As in the original contract, the fee included in the extended contract, which will run until June 30, 1956, is $1 above and beyond the costs which are incurred by G.E. in carrying out its contract and which are reimbursed from government funds. The contract extension also provides for renegotiation of the $1 fee at some time in the future if this seems desirable.

G.E. is making a refund to the government in the amount of approximately $10,000,000 for unexpended funds advanced to the company under the original contract. The extended contract calls for monthly advances to G.E. of $100,000 for administrative costs and for refunds of any unexpended portions of the monthly advance on an annual basis in the future. Twice that amount was advanced monthly under the original contract and was reduced for the new contract when experience indicated that actual administrative costs were appreciably less than originally estimated.

The old contract called for G.E. to have responsibility for both construction and operation. Under the new contract, G.E. will be responsible for operation only, with the AEC taking over construction in accordance with a pattern recently established at several other installations of the AEC.

G.E. will continue to manage the government-owned town of Richland, Wash., under the terms of the new contract.

BATTELLE SETS UP EUROPEAN RESEARCH INSTITUTE

A new research institute, Battelle International, will be established in Europe this fall, with financial support coming from industrial sponsorship, both European and American. It will be sponsored by Battelle Memorial Institute of Columbus, O., one of the world's largest industrial research foundations.

Maintaining laboratories and offices in several Western European countries, Battelle International will carry on investigations in many fields of research in existing research institutes, universities, and technical schools of Europe. Projects are planned, for instance, in the laboratories of such organizations as the Fulmer Research Institute (Great Britain), the TNO Council (The Netherlands), and the Max Planck Gesellschaft (Germany).

AEC GIVES STUDY CONTRACTS TO BENDIX AND TRACERLAB

Acceptance of two proposals to study the commercial feasibility of manufacturing, processing, and selling radioisotopes has resulted in the signing of Atomic Energy Commission contracts with Bendix Aviation Corporation of Detroit, Mich., and Tracerlab, Inc., Boston, Mass. These firms will carry on the studies at their own expense and must make their reports and evaluations directly to the AEC within a year.

The first step in the study will be an examination of the AEC's isotope processing program and an investigation into the possibilities of building and operating reactors to produce radioisotopes. Scientific staff members of the AEC and its contractors will assist the firms in their studies.

AMERICAN CYANAMID SIGNS TWO CONTRACTS FOR AEC WORK

It was recently announced that the American Cyanamid Company, New York, has signed two contracts with the Atomic Energy Commission for work connected with reactor fuel elements.

Under one contract, the company has agreed to take on operation of the chemical processing plant at the AEC's National Reactor Testing Station in Idaho Falls, Idaho. The plant, at

present under construction by the Bechtel Corporation, San Francisco, will recover nuclear fuel from used reactor fuel elements.

Terms of the other contract call for Cyanamid to operate a mineral dressing laboratory at Watertown, Mass., and to carry on research to find new uranium-recovery techniques.

KELLEX CHANGES NAME TO VITRO CORP. OF AMERICA

The Kellex Corporation, which engineered the Oak Ridge gaseous-diffusion plant, revealed last month that it has changed its name to Vitro Corporation of America. The change became effective August 31.

The corporation's organization, policies, and services will continue as before, it was stated by Albert L. Baker, president, who announced the change.

Since the war, the corporation, which was organized in January, 1943, as a subsidiary of the M. W. Kellogg Company, has designed major production facilities at Hanford, Wash., and has been engaged in several projects for the Army, Navy, and Air Force. The Vitro Manufacturing Company of Pittsburgh, Pa., purchased the firm in the spring of 1950.

The main offices of Vitro Corporation of America will remain at 233 Broadway, New York. The corporation operates research and development laboratories in West Orange and Jersey City, N. J., and in Silver Spring, Md.

REACTOR SCHOOL GRADUATES 46 STUDENTS, NEW TERM STARTS

A total of 46 engineers and scientists recently completed work in the 1950–51 session of the AEC's School of Reactor Technology at Oak Ridge, Tenn., and 23 of these students have already returned to the nine organizations which sponsored their training. The other 23 have accepted new positions with nine other industrial companies and government organizations.

Industrial companies employing the graduates include North American Aviation, General Electric Co., Phillips Petroleum Co., Consolidated-Vultee Aircraft Corp., Monsanto Chemical Co., E. I. du Pont de Nemours & Co., Pratt & Whitney Aircraft, Electric Boat Co., Bendix Aviation Co., and Westinghouse Electric Corp. Other graduates will work in AEC facilities in the Oak Ridge National Laboratory, Brookhaven National Laboratory, and AEC operations offices in New York and Chicago. The U. S. Air Force and USAF Air Materiel Command, the Bureau of Aeronautics, and the Bureau of Ships have taken the services of 16 of the graduates.

The 1951–52 term of the Reactor School is already under way, with an enrollment of about 70 students. Training facilities at the school have been expanded, and a new student laboratory is nearing completion.

OAK RIDGE TO OFFER COURSE ON HIGH-SPEED COMPUTERS

Oak Ridge Institute of Nuclear Studies, in cooperation with the Oak Ridge National Laboratory, will offer a course from December 3–14, 1951, in the techniques of preparing problems for high-speed digital computing machinery.

The course, to be given at Oak Ridge, will be centered around the electronic automatic computer, single address type, to be installed at ORNL, although modifications of machines and preparation techniques will also be considered.

A registration fee of $25 will be charged, and the course will be limited to 30 participants. Application blanks and additional information may be obtained from Ralph T. Overman, Chairman, Special Training Division, Oak Ridge Institute of Nuclear Studies, Oak Ridge, Tenn.

ORNL's Mathematics Panel Chief, Alston S. Householder, will be the

course director. He will be assisted by staff members of Argonne National Laboratory, Los Alamos Scientific Laboratory, the Computer Branch of ONR, and the Institute for Advanced Study.

IN BRIEF

➢***Uranium prospecting*** *is the subject of a best seller. Over 77,000 copies of the AEC's booklet "Prospecting for Uranium" have been bought since it was published in 1949. The manual is currently undergoing revision.*

➢**1,250,000 cubic yards of concrete** will go into construction of the AEC's Savanna River plant near Augusta, Ga. The Kolinski Concrete Company of Milwaukee, Wisc., "will pour." The company, which has a $15-million contract for the work at the Commission's production-center site, estimates the job will take three years.

➢***Fire*** *recently wrecked a secret laboratory of the uranium processing plant operated by the Vanadium Corporation of America in Durango, Colo. Damage was estimated at $25,000.*

➢**Proposed amendments** to the Atomic Energy Act, according to a recent statement of AEC chairman Gordon Dean, are presently "going the rounds of the Executive agencies, through the channels of the Bureau of the Budget, Defense, State, and so forth." Declining to elaborate on the contents of the amendments, Mr. Dean said that there is still a possibility of getting the changer through this session of Congress "if the Executive agencies will hurry up and give us their views."

➢***Autonomous atom towns*** *are "patently desirable," according to recommendations recently made to the AEC by the Panel on Community Operations which, for the past year, has been studying the problems of self-government and private ownership of real property in the Commission-owned towns of Oak Ridge, Tenn., and Richland, Wash. The recommendations, contained in a report issued by the Panel, are currently being studied by the residents of these communities who will attempt to arrive at a workable plan of self government supported by the AEC through Federal legislation.*

➢**Brazil** has taken steps toward installing a pilot atomic power plant. Announcement of this development was made last month by Alberto da Motta, president of Brazil's National Research Council and member of the United Nations Atomic Energy Commission since 1946. To be erected in Minas Geraes State, the plant, Admiral da Motta said, "will permit us to experiment on a reduced scale toward effective harnessing of nuclear energy. . . . Production of atom bombs in Brazil is not under consideration."

➢***Uniform security measures*** *for safeguarding atomic-energy information held in common by the United States, the United Kingdom, and Canada were sought in discussions at the fifth International Declassification Conference held last month in Washington. The recommendations resulting from the discussions will eventually result in revisions to the uniform Declassification Guides which the three nations use. Heading the American delegation was Dr. James G. Beckerley, director of classification of the U. S. Atomic Energy Commission. Sir John Cockcroft, director of the Atomic Energy Research Establishment at Harwell, headed the six-man British delegation, and Dr. W. R. Lewis was spokesman for the three Canadian delegates.*

➢**Norway's heavy-water reactor** was started up in initial tests at 100 kw early in August. The power level will probably be raised shortly to 300 kw. The neutron flux is expected to reach 3×10^{11} n/cm²/sec. Although Nor-

way built the pile (NU, June '51, p. 45), the Netherlands is supplying the uranium to operate it, and the two nations, through their national research councils, are exercising joint supervision over its operation. (An article on the reactor's construction will appear shortly in NUCLEONICS.)

➤ ***The 277 AEC patents*** *available on a royalty-free, nonexclusive basis, were met with increasing interest from American industry during the first half of 1951. From early 1949 to mid-1951, about 125 licenses to use these patents were granted, almost one half of which were issued during the first six months of this year. The patents cover, in general, the fields of general chemistry, uranium chemistry, electronic devices, and radiation-detection apparatus. Industries with serious corrosion problems, for instance, have shown interest in the patents on fluorocarbon and fluorolube production.*

NUCLEAR NEWSMAKERS

Bernard Peters resigned on September 1 as assistant professor of physics at the University of Rochester and is being succeeded by **Morton F. Kaplon.** Dr. Peters left the United States last month to accept a special chair in cosmic-ray research and teaching at Tata Institute in Bombay, India. Last year he spent four months at the institute on a cosmic-ray research project sponsored by the Atomic Energy Commission and the University of Rochester. Dr. Kaplon, who was co-discoverer in 1949 of the famous R-star (R for Rochester) with Dr. Peters and the late Helmut Bradt, has been placed in charge of the university's Cosmic Ray Group.

Forrest Western, formerly associate director of the Health Physics Division of Oak Ridge National Laboratory has taken up a new post in Washington in the AEC's Division of Biology and Medicine, Biophysics Branch.

Reorganization of the General Electric Company's Hanford Works Radiological Sciences Department, which is made up of three sections, Biophysics, Biology, and Radiological Records and Standards, became effective last month. **Herbert M. Parker,** director of the new department (formerly called the Health Instruments Division) announced the following appointments:

M. L. Mickleson is named assistant to the director, a continuation of his former position.

C. C. Gamertsfelder, named manager of the Biophysics Section, has a dual responsibility as head of Radiation Measurement. Under the Biophysics Section are also the Control Unit with **R. C. Thorburn** as head, and Environmental Hazards and General Studies Unit with **J. W. Healy** as head.

H. A. Kornberg is named manager of the Biology Section, with **R. F. Foster** as head, Aquatic Biology Unit; **R. C. Thompson** as head, Metabolism Unit; and **L. K. Bustad** as head, Toxicology Unit.

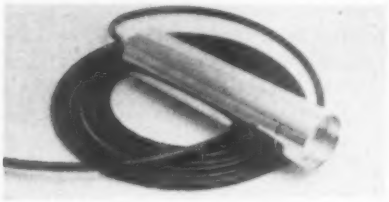
W. A. McAdams is named manager of Radiological Records and Standards Section. **H. A. Meloeny** is head, Exposure Records Unit; **J. M. Smith** is head, Standards Unit; **A. J. Stevens** is head, Radiation Monitoring Services Unit; and **G. L. Helgeson** is supervisor, Calibrations, all under Radiological Records and Standards Section.

Urner Liddel, who headed the Nuclear Physics Branch of Navy's Office of Naval Research, has been appointed Director of Physical Sciences. This division within the ONR includes three branches—physics, nuclear physics and electronics.

Jorgen Koch, assistant to Niels Bohr and official of the Institute of Theoretical Physics in Copenhagen, arrived in this country recently. He plans to

(Continued on page 89)

PRODUCTS and MATERIALS

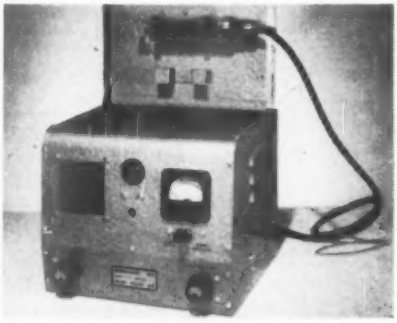


SCINTILLATION COUNTER

Nuclear Research and Development, Inc., 1094 Sutter Ave., St. Louis 5, Mo. The model SC-2 gamma end-window scintillation counter uses a NaI(Tl) activated crystal mounted in a polystyrene holder to provide maximum optical transmission. The unit incorporates a 5819 photomultiplier tube, and is housed in a chrome-plated brass cylinder. The necessary dropping resistors are mounted on the base of the 5819 tube socket. A cathode follower preamplifier circuit may be installed, if desired. The counter comes complete with cable, ready to be used in conjunction with any commercial scaler or count-rate meter.

TIME-DELAY GENERATOR

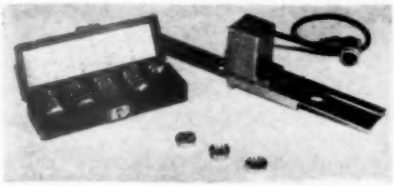
Rutherford Electronics Co., 3707 So. Robertson Blvd., Culver City, Cal. The model A-4 time-delay generator is a device for producing variable time delays ranging from 10 μsec to 10 sec. Five delay ranges, in decade steps, are provided, giving a full-scale reading of 0.001 sec delay on the lowest range. Blocking-oscillator and single-shot-vibrator output wave forms are provided. The pulse from the blocking oscillator is said to be 0.5 μsec wide with a rise time of 0.1 μsec. The pulse from the multivibrator is presented through a low-impedance cathode follower for easy viewing on the screen of a synchroscope; its width is a function of the delay range. The linearity of the instrument is said to be 0.3% between 0.1 scale and full scale; long-term accuracy is 1%. A calibrator assembly to permit checking of the calibration accuracy is available.



DEMONSTRATORS

Nuclear Research Corp., 2707 Federal St., Philadelphia 46, Pa. The models DU-1A and DU-2A are designed to demonstrate such things as particle detection, properties of absorbers, properties of G-M tubes, and the statistical nature of nuclear radiation. Both instruments will also serve as laboratory

monitors and, with suitable calibration, as survey meters. The model DU-1A does not have a built-in voltmeter. In both instruments the voltage is variable from 100–1,000 volts.



ABSORBER CHANGER

Tracerlab, Inc., 130 High St., Boston 10, Mass. This automatic absorber changer is an attachment for use with the concern's automatic sample changer. It consists of a slide and switch assembly and a set of 25 calibrated aluminum absorbers. When mounted, the device automatically positions the absorbers over the source. The absorbers, ranging in value from 1.6 mg/cm² to 1.5 gm/cm², are individually weighed; the weight-per-unit-area calibration is said to be accurate to 0.1%. The aluminum foil absorbers are 1 in. in diameter and are cemented in aluminum rings.

AIR FILTER

Cambridge Corp., 350 South Geddes St., Syracuse, N. Y. These filters, made of a soft paper that contains asbestos fibers, will remove 99.98% of all foreign matter in the air, according to the manufacturer. The filter is said to work well for particles in the size range 0.2–0.3 μ in diameter. Two sizes are available; the larger is rated at 850 ft³/min with dimensions of 24 × 24 × 11½ in.; the smaller is rated at 500 ft³/min with dimensions of 24 × 24 × 5⅞ in. When new, the static-pressure resistance of the filter is less than 1 in. of water.

SURVEY METER

Nucleonic Corporation of America, 497 Union St., Brooklyn 31, N. Y. The model RM-2 beta-gamma monitor has ranges of 0.5, 5, and 50 mr/hr, full scale, for the measurement of dosage rate. For particle detection it has ranges of 500, 5,000, and 50,000 cpm, full scale. The probe design permits differentiation between beta and gamma radiation. Accuracy is said to be 1% at high counting rates. The high-voltage supply is regulated at 900 volts. Modifications are available which have, in addition, a supply that is variable from 300 to 1,600 volts, and provision for monitoring alphas.

PORTABLE OVEN

Grieve-Hendry Co., Inc., 1101 N. Paulina St., Chicago 22, Ill. The model PL-1 portable electric oven has room for eight 2 × 12 × 23-in. drying pans. Operating on 110 volts the oven is capable of heating to 225° F in 15 min. Construction is all steel, with air-cell asbestos insulation. Forced air circulation provides uniform temperature throughout. A motor driven fan draws in fresh air and exhausts stale air, thus avoiding stratification and

making the oven practical for any dehydration or baking process in the laboratory.

PRESSUREGRAPH

Electro Products Laboratories, 4501 North Ravenswood Ave., Chicago 40, Ill. This device is designed to record pressure–time measurements of static, dynamic, and transient pressures in air, gases, water, and viscous liquids. The pressure range covered is 0.5–10,000 lbs/in^2; frequency response ranges from 0 to 20,000 cps; accuracy is said to be 1% of full scale or rated pressure. The linearity of the instruments varies slightly with diaphragm thickness. The electronic pickup is connected to the region where pressure is to be measured; the remainder of the instrument can be placed as far as 200 ft away. The temperature of the pickup must be held below 250° F; facilities for water or air cooling are available.

PLASTIC CONTAINERS

American Agile Corp., 5806 Hough Ave., Cleveland 3, O. Bottles, jugs, and jars fabricated of polyethylene are available in capacities ranging from 1 quart to 5 gallons. Of completely rigid construction, these containers are said to be shockproof and unbreakable at temperatures from sub-zero to 180° F. Besides being very light in weight, the containers resist the actions of most acids and alkalies, including sulfuric and hydrofluoric acids.

MERGER

Georator Corp., Arlington 9, Va. has acquired the complete business and manufacturing facilities of the **Geophysical Instrument Co.**, also of Arlington. Manufacture of well survey instruments, including special radiation detectors, will be continued.

LITERATURE AVAILABLE

Lignum-vitae. Bulletin describes properties and uses of this very hard, acid-resistant wood. *Lignum-Vitae Products Corporation, 10 Boyd Ave., Jersey City, N. J.*

Pressure transmitters. Engineering-data folders illustrate and describe complete line of pressure-measurement transmitters. *Automatic Temperature Control Co., Inc., 5200 Pulaski Ave., Philadelphia 44, Pa.*

Dense glass. Circular GS-3 gives information on glass for radiation-shielded viewing. *Penberthy Instrument Co., Inc., Hi-D Division, 666 Adams St. Seattle 8, Wash.*

Wire strainers. Manual illustrates design of wire-cloth strainers. *Michigan Wire Cloth Co., 2100 Howard St., Detroit 16, Mich.*

Electronic tubes. Catalog lists line of tubes available. *Milo Radio and Electronics Corp., 200 Greenwich St., New York 7, N. Y.*

Arctangent paper. Bulletin describes uses of arctangent coordinate graph paper. *Orbit Electric Co., 2710 N. Menard Ave., Chicago 39, Ill.*

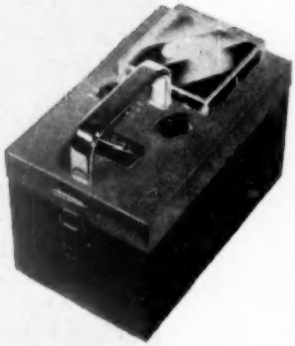
Electrical measurements. Booklet B-4696 tells "how to select" electrical instruments. *Westinghouse Electric Corp., Box 2099, Pittsburgh 30, Pa.*

Vacuum equipment. Catalog 11K lists line of high-vacuum equipment. *Research Vacuum Supply Co., 3434 W. Montrose Ave., Chicago 18, Ill.*

Metallograph. Folder describes device for examining and making microphotographs of metal. *F. T. Griswold Mfg. Co., 305 W. Lancaster Ave., Wayne, Pa.*

G-M Tubes. Illustrated folder describes counter tubes. *20th Century Electronics, Ltd., Dunbar Works, Dunbar St., W. Norwood, London, SE 27, Eng.*

TWO NEW SURVEY INSTRUMENTS BUILT BY EL-TRONICS



Ionization Chamber Type Survey Meter

Survey Meter PR-50 combines a light weight (8 pounds) with rugged, dependable durability. Five individual ranges in distinctive colors read from 0 to 50,000 MR/HR on a large Navy sliding scale meter. Meter scale changes with range switch. Rugged weatherproof construction. Battery life of 1000 hours and provision for shoulder strap make this model a versatile, portable instrument.

Beta-Gamma Survey Meter ("Cutie Pie")

This miniaturized light weight instrument has a range of 0 to 2500 MR/HR. Splash-proof design. Calibration accuracy ±10%. Battery life 1000 hours. A fine hand survey instrument of unusual accuracy and excellent stability. Rotating beta-gamma shield on front of chamber.

WRITE FOR COMPLETE SPECIFICATIONS

El-Tronics INC.

2647–67 N. Howard Street
Phila. 33, Pa.

committee studying energy resources said that, "Isolated and somewhat spasmodic attempts have been made to utilize direct solar radiation by means of solar boilers and by means of the so-called heat pump."

Widespread use of the heat pump and direct collection of solar energy for comfort heating has been limited by cost and geographical considerations (availability of sunlight).

Most interesting as far as solar energy is concerned is controlled biological photosynthesis. Some work is being done on *Chlorella*, a single-celled organism which absorbs energy throughout the visible spectrum. However, the lowest over-all costs now in sight are 10–50 times coal costs and, unless something startlingly new is uncovered, it will not be possible for organic photosynthetic fuels to compete with present costs of coal.

But our purpose here is not to belittle the possibilities of developing solar energy. Rather it's to show that it is still an unknown, as is nuclear energy.

And what about nuclear energy?

Nuclear Energy

We are firmly convinced that it is much too early to make any valid analysis of the role which nuclear energy will play in our industrial economy. We are reminded here of the very pessimistic predictions made by responsible people about the future of such things as the gas turbine, jet propulsion and even the airplane.

The atomic energy field is in its infancy. Much theoretical work has been done. Some reactors have been built for use in plutonium production and in research. Much has been said about nuclear power. But the fact remains that we do not yet have an operating nuclear power plant!

Technological advance comes from experimentation, from doing. It is only in this way that we become intimate with our problems, find out what their true nature is.

There is no question but that the obstacles which stand in the way of economically feasible nuclear power are tremendous. But we have hardly scratched the surface in our attempts to overcome these obstacles. This is emphasized by one AEC official who says, "Because this is a relatively young field, the opportunity for technical improvements which will effect major economies is still great, as in most new industries. . . . At this early stage, it is certainly unwise to lose faith in the capabilities of our sciences for achieving the necessary economies and to relegate the future of nuclear power production only to isolated spots on the basis of relative ease of transportation of nuclear fuels."

An important factor in the future utilization of nuclear energy is the availability of fissionable material. If breeding is successful, it has been predicted that the recoverable heat content of the world's reserve of uranium and thorium would yield about $1{,}700 \times 10^{18}$ BTU. A considerable part of this might be available at a cost of not more than $100 a pound; and with improved techniques, perhaps 100×10^{18} BTU might be available at $50 a pound, a very favorable price.

Compared with the figure of 30×10^{18} BTU for the world energy reserves

(exclusive of nuclear and solar energy), it may be possible for a period of several hundred years for nuclear energy to carry up to 20% of the total load in the world energy system. Even if the figure were as low as 5 or 10%, this would be substantial, especially when one realizes that our hydroelectric power system today contributes only 1% and is still a robust industry.

Waste Disposal

Another obstacle in the development of nuclear power, according to Dr. Conant, is the problem of the disposal of radioactive wastes from power reactors. Here we must repeat that we don't know enough yet. Disposal of wastes is very costly, dangerous, and space-consuming. However, work is now being done, particularly at Chalk River, on the complete utilization of these wastes, both the radioactive and nonradioactive constituents.

We may never reach a nuclear millennium, but let's by all means study the situation intensively—both nuclear and solar energy. Actually, it was only recently that we had the first substantial show of industrial interest in the atomic energy field. As pointed out previously (NU, July '51, p. 2), by next year the reports of the eight companies studying the feasibility of building power-producing reactors may give us the first clear picture of the future of this industry that we have yet had.

Only when our full technological resources have been applied to the problem will we come up with an answer. —*J.D.L.*

Nucleonic Events *(Continued from page 83)*

spend a year at Massachusetts Institute of Technology doing fundamental physics research.

H. McManus, former lecturer in mathematical physics at the University of Birmingham, England, has accepted appointment as research officer at the Canadian Atomic Energy Project in Chalk River, Ont.

Philip N. Powers has become executive administrator of the power pile project recently initiated by the Monsanto Chemical Company, St. Louis, Mo. Previously associated with the AEC, he resigned as manpower consultant with the NSRB to take the Monsanto post.

Donald D. Cowen, who has been associated with work at Oak Ridge since 1943, has been named superintendent of the Information and Reports Division, a new division at Oak Ridge National Laboratory. Mr. Cowen held a similar position with the Fairchild Engine and Airplane Corporation in connection with the now-defunct NEPA project.

Patrick J. Selak, formerly on the staff of the office of research and medicine, AEC Oak Ridge Operations, has accepted the post of executive officer to the director of the technical and production division, AEC Savannah River Operations.

Robert L. Corsbie has been appointed chief of the Civil Defense Liaison Branch of the AEC's Division of Biology and Medicine. He will be responsible for liaison with the Federal Civil Defense Administration and other agencies in connection with protection against hazards associated with atomic

energy and methods for building structures to minimize radiation and blast effects of atomic weapons. **Harry L. Bowman** had been serving as acting chief. As head of the Department of Civil Engineering, Drexel Institute, Philadelphia, Professor Bowman will continue to serve as a consultant to the division.

Lieut. Gen. Elwood R. Quesada, who just completed a two-year special assignment as commander of Joint Task Force Three which conducted the recent AEC weapons tests at Eniwetok, has retired from the Army after 26 years of military service.

Irving Tannenbaum, John Beyster, Max Walske, Jr., and **James Terrell Jr.,** have joined the staff of the Los Alamos Scientific Laboratory. Dr. Tannenbaum is working as a member of the Chemistry and Metallurgy Division, and Drs. Beyster and Terrell are with the Experimental Physics Division. Dr. Walske, who was employed at the Laboratory in 1947, this year received his doctorate from Cornell University and is connected with the Theoretical Physics Division.

Lt. Gen. Sir Frederick E. Morgan has been named Controller of Atomic Energy to head Great Britain's atomic energy program. The Ministry of Supply selected him as successor to Lord Portal, whose resignation from the post became effective on October 1 (NU, Aug. '51, p. 81).

Manson Benedict, on leave from Massachusetts Institute of Technology as professor of nuclear engineering, has taken up assignment as a special technical assistant to the general manager of the Atomic Energy Commission. His task is to organize an operations analysis staff for the AEC in connection with solution of its technical problems.

Richard Stokes has accepted a position at Los Alamos Scientific Laboratory. He recently received his doctorate at Iowa State College where he was associated with the Institute for Atomic Research.

DeWitt Clinton Hamilton, Jr., formerly assistant professor of mechanical engineering at Purdue University, has joined the staff of the Reactor Technology Division of Oak Ridge National Laboratory. **Arthur Canfield Upton** has joined ORNL's Biology Division. Dr. Upton previously taught pathology at the University of Michigan.

MEETINGS

National Electronics Conference—Chicago, Ill., Oct. 22–24

American Institute of Physics in joint meeting with Founding Societies of the Institute (American Physical Society; Optical Society of America; Acoustical Society of America; Society of Rheology; American Association of Physics Teachers; also American Crystallographic Association)—Hotel Sherman, Chicago, Ill., Oct. 23–27

American Physical Society in joint meeting with American Institute of Physics—Chicago, Ill., Oct. 25–27

Institute of Mathematical Statistics—Washington, D. C., Oct. 26–27

American Mathematical Society—Washington, D. C., Oct. 27

American Institute of Mining and Metallurgical Engineers—Mexico City, Mex., Oct. 28–Nov. 3

American Physical Society, Division of Electron Physics—Washington, D. C., Nov. 1–3

National Academy of Sciences, Autumn Meeting—Yale University, New Haven, Conn., Nov. 5–7

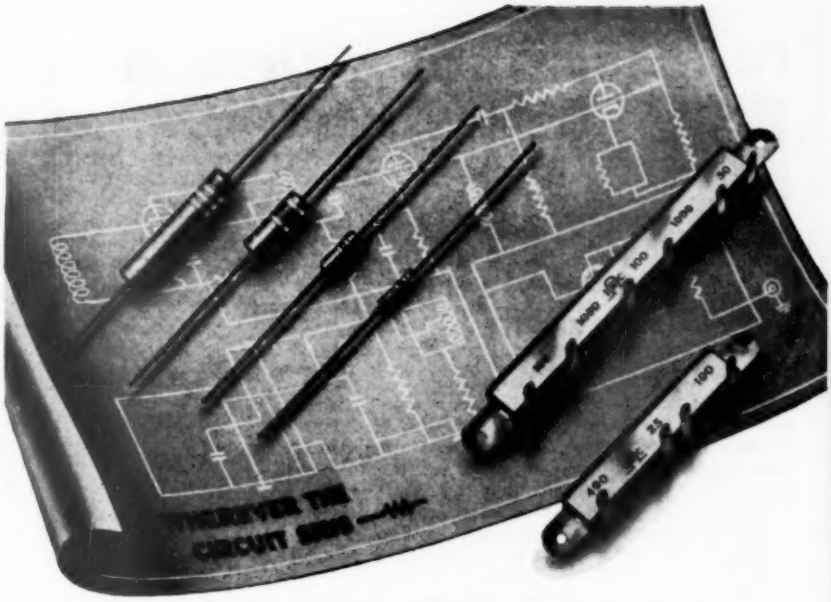
International Symposium on Abrasion and Wear—Delft, Holland, Nov. 14–15

Society of Exploration Geophysicists—Baker Hotel, Dallas, Tex., Nov. 19–20

American Physical Society—Houston, Texas, Nov. 23–24

American Society of Mechanical Engineers, Annual Meeting—Chalfonte-Haddon Hall, Atlantic City, N. J., Nov. 25–Dec. 1

Conference on Laboratory Design for Handling Radioactive Materials, sponsored by American Institute of Architects and Atomic Energy Commission—National Academy of Sciences, Washington, D. C., Nov. 27–28



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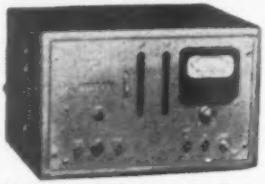

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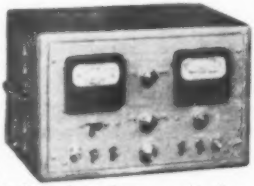

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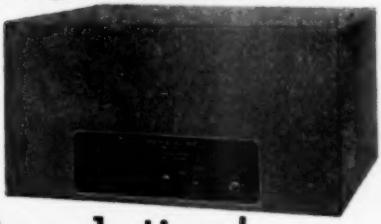

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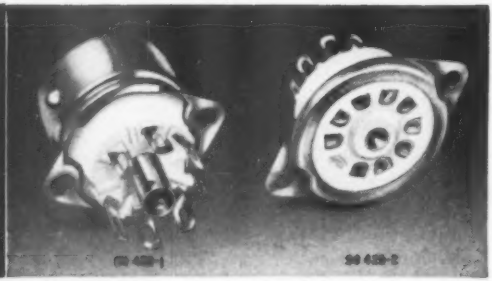

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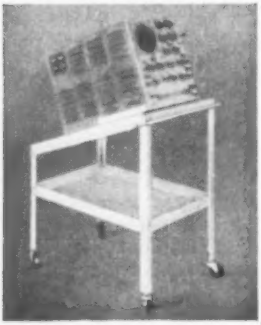

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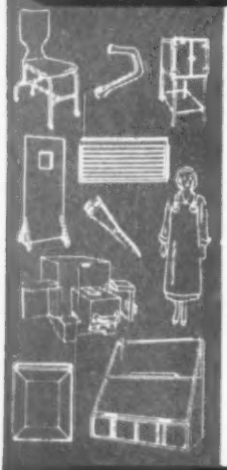

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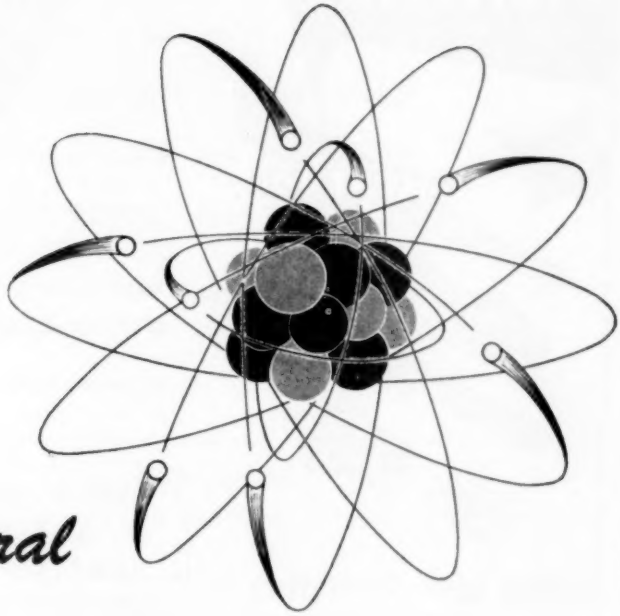

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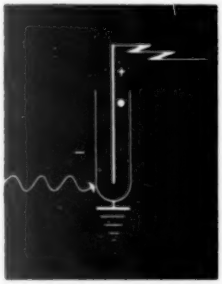

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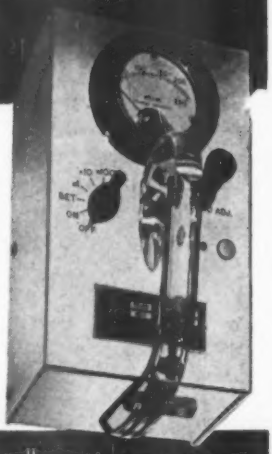

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INDEX TO ADVERTISERS

OCT. 1951

American Phenolic Corporation 100
Bar-Ray Products, Inc. 98
Berkeley Scientific Company 92
Bersworth Chemical Co. 100
Boeing Airplane Company 103
Eldorado Mining and Refining (1944) Limited 99
El-Tronics, Inc. 87
General Electric Co. 4
International Resistance Co. 91
Kellex Corporation 100
Kewaunee Mfg. Co. . . . . Third Cover
Leitz, Inc., E. 97
Little, Inc., Arthur D. 98
McGraw-Hill Book Co. 94
Nuclear Instrument & Chemical Corporation . . . . . Fourth Cover
Nuclear Research Corp. 102
Ohio Mica Film Co. 104
Ohmart Corporation 101
Searchlight Section 104
Sorensen & Co., Inc. 93
Technical Associates 102
Technical Service Corp. 96
Tracerlab, Inc. 95
United States Gasket Co. 96
Victoreen Instrument Co. . . . Second Cover
Vitro Corporation of America 100

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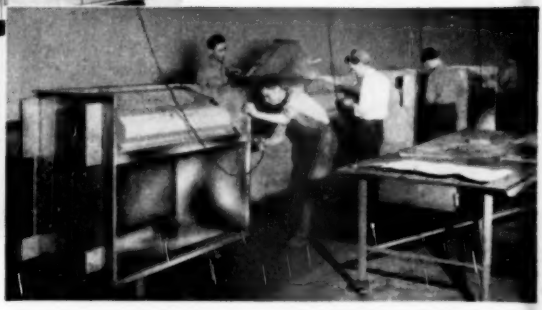

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